Synthesis of Optimal Heat and Mass Exchange Networks using a Two-Step Hybrid Approach Including Detailed Unit Designs

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Abstract

Heat exchanger network synthesis (HENS) and mass exchanger synthesis (MENS) have been studied extensively. By recovering excess heat and mass from within a process it is possible to drastically reduce raw material and energy usage; increasing overall network efficiency and profitability, and reducing the environmental burden of a process. Modern approaches to the MENS and HENS problem have involved formulating the problem as a mixed-integer nonlinear program (MINLP) through the use of superstructures, wherein many of the possible network topologies are embedded. These large MINLPs can be difficult to solve to global optimality due to the non-convex formulation required and thus current MINLP-based superstructures make use of simplified shortcut models to represent the individual units.

Something that has yet to be adequately addressed in the literature is the fact that these simplified MINLP formulations result in networks having individual units that can be shown to be very different to rigorously designed units. This often means that an optimal solution from the MINLP network optimisation may not be the true optimum once the individual units are rigorously designed. This thesis presents a novel approach for the design of optimal HENs and MENs through a hybrid design method that makes use of both an MINLP formulation for the network topology optimisation and a rigorous design procedure for the individual units. The method utilises a number of correction factors that update the MINLP solution network to more accurately represent the designs obtained from the rigorous designs so that information that is not explicitly available to the MINLP optimisation can be included. This allows for the MINLP optimisation to be guided to more realistic solutions by information from the detailed unit designs.

The fact that the newly developed method's MINLP formulation does not include any additional nonlinearities is vital as, even with the current state-of-the-art formulations and solvers, it is still a major challenge to find globally optimal solutions; especially for larger problems. The method, by making use of a number of iterations with small incremental changes to the correction factors between successive iterations, is shown to be very effective at generating large numbers of candidate networks. These networks do not only look more like the actual networks that could be obtained in a real design, but also have new initial points at each iteration. Repeated use of systematically obtained initial points, in what is termed a multi-start approach, can be seen to be extremely useful in increasing the

chances to obtain a globally optimal solution, especially for problems that involve many networks with similar costs, but different topologies.

A further advantage of the method is that a variety of techniques can potentially be used for the detailed unit designs and new information that has not previously been considered in the MINLP network synthesis can be included to guide the network topology, without increasing the formulation's non-convexity. This is demonstrated in the thesis by using different approaches for each of the chapters. In Chapter 4, an industry-standard heuristic approach is used for the individual heat exchanger designs; with the number of shells, changes related to TEMA decisions, pressure drops, and different overall heat transfer coefficients for each exchanger all being included implicitly. Chapter 5 shows how the new method can be extended to multiple periods of operations in HENS, which further shows that even more information can be included to guide the designs without increasing the complexity of the MINLP. This is particularly important for multi-period problems that can be extremely large and therefore difficult to solve to global optimality. Chapter 5 also shows that detailed design decisions, that involve judgement from the designer, can be included, such as the tolerated over-design of a unit and whether additional units are necessary to perform a specific task across multiple operating conditions. Chapter 6 shows how a detailed nonlinear program (NLP) unit optimisation can be included that allows for automated detailed network generation. In this chapter the methodology is applied to MENS, with the detailed unit designs performed automatically through the development of a novel NLP formulation that finds the optimal packed columns based on the mass balances from the MINLP using orthogonal collocation on finite elements (OCFE) to solve the differential equations involved. It also involves a novel approach to including flooding considerations and the optimal packing sizes which has erstwhile been ignored in MENS approaches.

The thesis lays the foundation to including detailed designs into MINLP superstructure-based optimisation and is demonstrated at being effective at generating many potential networks that are highly competitive with existing solutions to case study problems, while also ensuring that the networks generated are applicable to industrial settings.

Declaration

I, Michael Short, hereby declare that the work on which this dissertation/thesis is based is my original work (except where acknowledgements indicate otherwise) and that neither the whole work nor any part of it has been, is being, or is to be submitted for another degree in this or any other university.

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Michael Short

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Chapter 1

Introduction

Chapter 1: Introduction

The chemical industry has increasingly shifted its focus to finding ways in which to minimise raw material usage, energy, and emissions over the past 40 years as a result of the increased costs of raw materials and energy, as well as growing socio-political pressure to reduce human impact on the environment. Most chemical industrial research has focused on the development of specific technologies that allow for the production of products that meet specifications determined by the market. This has resulted in an appreciable amount of valuable knowledge into the details of operation and design for many unit operations that convert raw materials into these products such as reactors, separators, heaters, dryers, coolers, etc. Equally important in modern chemical plants is the need for these individual process elements to be thought of as part of a larger integrated process. With design decisions in one area of the plant directly and indirectly having effects on other areas, it is an extremely challenging task for a designer to find optimal designs for large plants where variables in process operating conditions such as pressures, temperatures, flowrates, etc. can run into the thousands. Process synthesis can be defined as the general methodology used to find the best combinations of technologies and operating conditions needed in order to have the best-functioning plant in terms of specified objectives that include minimising external energy and raw material input, maximising product or profit, improving safety of operation, or minimising the environmental burden associated with a process.

1.1 Process Integration

Attempting to enumerate and discover the many complex relationships that exist within such large systems is a difficult task, even with the help of modern computational power. Most modern engineers attempt to design processes by first selecting well-understood conventional technologies for the reactor and separation stages and using a process simulator or design models to represent these. Around these key units the rest of the

process is designed to include the mixers, additional separators, process utilities, storage, etc. Once the key areas of the plant are designed the process engineer might look toward process integration techniques in order to save on costs.

The field of process integration and intensification focuses on methods to reduce the impact and cost of particular processes by finding ways to utilise waste heat or mass from within a process to fulfil the requirements for heat or mass in other areas of the process. Process integration attempts to link different areas of the plant together in order to create a holistic design that increases overall efficiency by exploiting the interactions between different units and can result in very large savings across the plant. In fact, effective process integration can often make the difference between two competing companies' processes that utilise similar technologies to create the same products.

The accepted standard method in industry described above has the significant disadvantage that each section of the plant is designed sequentially, with little regard for the impacts of the individual units' design on subsequent units, or their integration into the rest of the plant. Key areas of research within this field have been the optimisation of heat exchanger networks (HENs) and mass exchanger networks (MENs). A cornerstone development in the design of these systems is that of pinch technology (PT) (Linnhoff & Flower, 1978). PT allows the designer to set particular targets for the design, prior to any design decisions, through the enumeration of the physical and thermodynamic limits from the system in question. These targets are then used by the designer to develop a design that attempts to get as close to the target as possible. PT has the distinct disadvantage that the design evolves over time as the designer attempts to find a design that reaches the target as well as the fact that only a single target can be set and reached at a time. PT is described in more detail in Chapter 2.2.

Traditionally these sequential approaches have been effective in improving solutions, however a designers' judgement and intuition for more optimal designs can often lead to sub-optimal solutions, and therefore rigorous systematic approaches have been developed that have been shown to find significantly better solutions.

1.2 Mathematical Programming

Sequential design approaches, while still an industry standard and taught at the undergraduate level, cannot guarantee a globally optimal solution. With the rise of computational power and continuing developments in the field of optimisation mathematics, rigorous methods have been developed that allow for simultaneous optimisation approaches. In these methods the problem is described using a mathematical representation through the use of constraints for the physical properties and thermodynamics of the problem; where parameters include the process data and design decisions, such as flowrates, temperatures, unit sizes, etc., are included as variables. This formulation can then be optimised with regards to a desired objective function.

Mathematical programming has been used to automate sequential approaches in linear programming (LP) and mixed-integer linear programming (MILP) models for HENS (e.g. Papoulias and Grossmann, 1983) and MENS (e.g. El-Halwagi and Manousiouthakis, 1990a), however can be shown to be outperformed by simultaneous optimisation approaches (Yee and Grossmann, 1990a). In simultaneous approaches the HENS and MENS tasks are often placed into a superstructure that aims to embed as many possible network structures within it, and a nonlinear program (NLP) or mixed-integer nonlinear program (MINLP) is formulated that makes use of optimisation solvers to find an optimal network.

Modern NLP and MINLP solvers have seen significant advances in the last few decades with both deterministic and stochastic solvers seeing increased application. A detailed description of current techniques can be found in Chapter 2.4. A key problem that still exists however is the fact that for non-convex formulations there is no way to guarantee globally optimal solutions. Because of this key limitation, most MINLP and NLP approaches to HENS and MENS have focused on techniques to reduce non-convexity in order to find globally optimal solutions at the cost of model complexity. In reducing the nonlinearity of the topology formulation through the use of simplified models to represent the individual exchangers, it can be shown that the solutions generated are often quite different to those of a rigorously designed network with regards to network costs.

1.3 Motivation and Summary

A large body of research exists in applying MINLP optimisation techniques to the synthesis of both MENs and HENs. Due to the limitations of modern deterministic solvers, many of the attempts at formulating these problems have dealt with simplified representations of the individual exchangers. Even with these simplified representations it is not possible to formulate the problem as a convex problem if one hopes to include many potential networks within the superstructure, such as allowing for the possibility of stream splitting with non-isocompositional and non-isothermal mixing, as well as including detailed capital costing functions. This leads to the problem that it is impossible to guarantee a globally optimal solution, which becomes increasingly problematic as the size of the problem grows. These superstructure-based non-convex formulations can generate many different networks and embed a very large number of potential solutions within them. They are successful in determining, at least locally, optimal flowrates, mass balances, and heat balances, but they are limited by the simplified representations of the individual exchangers themselves. In order to avoid highly non-linear equations, as well as singularities, a large number of simplifications are made that ignore many key aspects of heat and mass exchanger design. As the key variable in many of these optimisation studies involves the capital costing of the network itself, these simplifications often result in networks that, while potentially optimal for the fixed parameters present in the mathematical representation, are not actually optimal once more detailed considerations are taken into account.

To date very little work has been done to address this problem, with work concentrating predominantly on either network design using simplified exchanger representations, or on the optimisation of singular units. This thesis presents a novel attempt to bridge the gap between these two very important topics in process synthesis in order to synthesise optimal MENs and HENs. The algorithm developed within this thesis uses a similar MINLP formulation to other authors to find the optimal network structure, however it uses an updated objective function that allows for more detailed aspects of the network to be included. These new inclusions, which are implicitly determined, can allow for aspects such as multiple shells, heat and mass transfer coefficients based on the specific exchanger designs involved in each match, pressure drops, etc. to be introduced without adding to the

nonlinearity of the problem. This is possible through the use of correction factors that are determined in a sub-optimisation step that involves the rigorous design of the network. The sub-optimisation step uses the topology, flowrates, mass balances, and heat balances determined by the MINLP to design the individual units using NLP optimisations, heuristics, or other software. The sizes and costs obtained from the rigorously designed and verified network are then used to derive correction factors that are inputted into a subsequent run of the MINLP. The correction factors are implemented in such a way so as to correct the simplified models of the MINLP to more accurately resemble the rigorous solutions and thus guide the MINLP to select a new topology based on the more accurate information. The iterative procedure iterates between the MINLP topology optimisation and the rigorous network design until either the correction factors converge to a solution and the objective functions of both the MINLP and rigorous network are identical or until a maximum number of iterations is reached.

The method has a number of advantages over conventional methods:

1. Each design is verified with detailed methods to guarantee that the designs can be implemented in real-world problems.
2. The optimal network is selected based on the detailed network design, not the simplified models of the MINLP, ensuring accuracy.
3. The MINLP has access to more detailed aspects of the design implicitly, meaning that the network can be designed with the optimiser having access to this information while not increasing the nonlinearity within the MINLP formulation.
4. There is a far greater chance that a globally optimal solution is obtained due to the systematic generation of new initial points and restarts of the MINLP due to the implementation of the correction factors at each iteration.

In addition to these general advantages, the application of the algorithm to specific examples of single period heat exchanger network synthesis, multi-period heat exchanger network synthesis, and mass exchanger network synthesis, shows that the method can easily be expanded to different applications with specific advantages in each case over traditional methods.

1.4 Scope and Structure of Thesis

This thesis will develop the method summarised above and apply the methodology to three separate applications. The applications have all been published or submitted for publication, and so are presented in full. The permission to include these publications within this thesis has been obtained. While there are co-authors on each of the papers and their contributions are acknowledged, each co-author acknowledges the current author as the sole worker in producing the studies, with each co-author providing feedback and valuable ideas as is typical in supervisory roles in academia. This thesis aims to show the merits and usefulness of the algorithm and how it can be extended to a number of applications.

Chapter 2 gives a comprehensive review of the pertinent literature and theory necessary in understanding the contributions of this thesis, as well as in placing the work in the field in general. Specifically, the literature review focuses on current optimisation techniques and their application to the field of network synthesis, with a special focus on HENs and MENs, as well as on the selection and design of the individual units within these networks.

Chapter 3 presents the proposed approach summarised above, and the general methodology that is applied to the subsequent applications. It summarises the philosophy of the approach in a general form, with its application detailed in the subsequent chapters.

Chapter 4 presents the application of the algorithm and design philosophy derived in Chapter 3 to the synthesis of optimal single period heat exchanger networks. This is done in the form of presenting a verbatim copy of the paper written by this author entitled "Synthesis of heat exchanger networks using mathematical programming and heuristics in a two-step optimisation procedure with detailed exchanger design" published in the 144th volume of *Chemical Engineering Science* Journal on 4 February 2016 (Short, et al., 2016a). It should be noted that, since this is a reproduction, that this chapter includes its own literature review, nomenclature, appendices, and references. It is meant to be accessible as a stand-alone version of the method presented in this thesis (as with Chapters 5 and 6), as well as to be read as a chapter in this thesis that shows the methods applicability to the particular problems therein.

Chapter 5 presents the algorithm's application to the quite different problem of multi-period HENS, wherein a far more comprehensive evaluation of the networks is required, as well as special additions to allow for the network to operate at different operating conditions. This chapter, like Chapter 4, is a reproduction of a paper entitled "Two-step hybrid approach for the synthesis of multi-period heat exchanger networks with detailed exchanger design" published in the 105th volume of *Applied Thermal Engineering* Journal on 17 May 2016 (Short et al., 2016b). It should be read with the same considerations as in Chapter 4.

Chapter 6 shows the method's applicability to the synthesis of optimal MENs. This problem, while it shares many commonalities with the HENs problem, has many significant differences that are outlined in Chapter 2, as well as in the chapter itself. This chapter is also presented as a copy of a paper entitled "Synthesis of mass exchanger networks in a two-step hybrid optimisation strategy", to be submitted to Chemical Engineering Science Journal. The paper extends the methodology to the case of MENS, while also introducing another novel contribution; the application of orthogonal collocation on finite elements to packed column design. This novel method, applied in the sub-optimisation of the method, allows for the simultaneous optimisation of multiple packed columns, including the selection of packing, optimal velocities and pressure drops based on flooding constraints, heights, diameters etc. in an NLP optimisation. This allows for a rigorous optimisation in the sub-optimisation, giving detailed solutions in a fast, robust, and reliable way, allowing for some degree of automation for the methodology as well. This chapter, presented in the format of a paper should also be read in the context of the overall methodology and thesis argument.

Finally Chapter 7 concludes the thesis by summarising the highlights and contributions of the work and presenting the recommendations and extensions of the design philosophy presented.

Chapter 2

Literature Review

Chapter 2: Literature Review

2.1 Introduction

The literature review presented in this chapter aims to acquaint the reader with the relevant literature in order to place this thesis within the context of previous work. This chapter will attempt to be comprehensive, however relevant literature for Chapters 4, 5, and 6, is also presented in those chapters because these chapters are reproductions of papers written by the author that apply the methodology to specific case studies. For this reason, the literature reviewed in this chapter will focus predominantly on the topics that are not covered in as much detail in Chapters 4, 5, and 6. This literature review will begin by reviewing modern techniques of process integration, specifically focusing on pinch technology and sequential mathematical programming approaches. Following this will be a review of mathematical programming techniques as applied to HENS and MENS, including a discussion of modern solver technology, with a specific focus on deterministic solvers in Section 2.4. Since the method developed in this study applies both rigorous models and network optimisation, a brief discussion on modern techniques to design individual heat exchangers and packed columns is also included, however much of this literature is also included in the relevant individual chapters and so will not be discussed in as much detail as the other topics.

As discussed in Chapter 1, a key aspect of modern chemical plants is process integration, with the majority of integration techniques focusing on HENs. HENs are designed in order to ensure that external energy usage within the plant is optimised by transferring heat from hot streams that need to be at lower temperatures to cold streams that require target temperatures that are hotter. This is achieved through the use of heat exchangers, process units that allow for the two streams to be passed by one another separated only by a thin metal wall, allowing for the heat to be transferred across the metal wall. The design of these devices will be described in Section 2.5.1, as well as in Chapters 4 and 5. In a large plant there can be many streams with various flowrates, compositions and supply and target

temperatures. If streams cannot reach their respective target temperatures through the use of streams already present within a process, utilities can be used. These utilities (normally steam or cooling water) often form a significant portion of the running costs of the plant, and therefore it is desired to find the optimal network of process to process stream matches to minimise the overall total annual cost (TAC).

In many process plants there exists a large number of process streams that require contaminants to be removed in order to reach a certain purity of product or in order to expel the stream into the environment, and a common method to remove contaminants involves the use of some kind of mass exchange operation. MENS can be defined in a similar way to HENS, with mass transfer being analogous to heat transfer in many ways. Mass exchangers are a broad category of operations that involve the transfer of a species from a rich stream (stream in which the species is in high concentration) to a lean stream or mass separating agent (MSA). This is analogous to hot streams and cold streams in HENs. In mass exchangers, similar to heat exchangers, the flow of mass is determined by a concentration gradient (analogous to a heat gradient) between the two streams. The individual units that can perform mass exchange operations include absorbers, strippers, adsorbers, ion exchangers, and extractors and usually work by passing two streams of different phases past each other in a way that attempts to maximise the interfacial/contact area. The design of these units is covered in more detail in Section 2.5.2 as well as in Chapter 6.

Throughout the literature review MENs and HENs will be discussed side-by-side as there are many parallels that can be drawn between these two seemingly different process integration tasks. While the individual unit design considerations involve vastly different approaches, the methods used to synthesise MENs and HENs are similar in many ways.

2.2 Pinch Technology on HENS

Systematic approaches to process integration were the result of intensified research into energy integration as concerns around energy security grew during the 1970s (Gunderson & Naess, 1988). There have been many different approaches to solving this problem, with over 400 papers being published in the last 50 years (as of 2002, so this number is probably up to well over 550 by now) (Furman & Sahinidis, 2002). One of the first systematic approaches

came with the invention of pinch technology (PT) by Hohmann (1971), although the use of the term "PT" was introduced by Linnhoff and Flower (1978). With external energy input reduction (minimal utility usage) through the use of heat exchanger networks as the primary goal, the method first identifies a minimum energy target. The energy target is determined by identifying the system's "pinch" points for a set value of the exchanger minimum approach temperature (EMAT or $\mathbf{\Delta}T_{min}$). These pinch points can be determined either through the use of a graphical representation via composite curve plots (Linnhoff, et al., 1982), or through some other algebraic method such as the problem table algorithm (PTA) (Linnhoff & Flower, 1978) or through an automated method using software. Once the pinch points are identified and the utility requirements fixed, the design can be divided into sections where heat exchanger placements can be decided, with the placement of heat exchangers across the pinch forbidden.

2.2.1 Minimum Utility Targets

The first target that was utilised in PT was for the minimisation of the external energy required for all the streams present to reach their desired target temperatures. The graphical approach used to identify this target requires the construction of composite curves and is the easiest way to demonstrate PT. In this method the axes of the two-dimensional plot are temperatures and cumulative enthalpy. The curves are constructed by first plotting the heat exchanged by each hot and each cold stream versus its temperature and then combining these into composite curves. In this representation the cold streams are represented by the cold composite curve (CCC) and the hot streams by the hot composite curve (HCC), where each point of inflection is a supply or target temperature of a stream with the slope of each line a representation of the combined streams' heat capacity flowrates that fall within that temperature range. The curves are then plotted onto the same set of axes, as shown in the example, Figure 2.1, below.

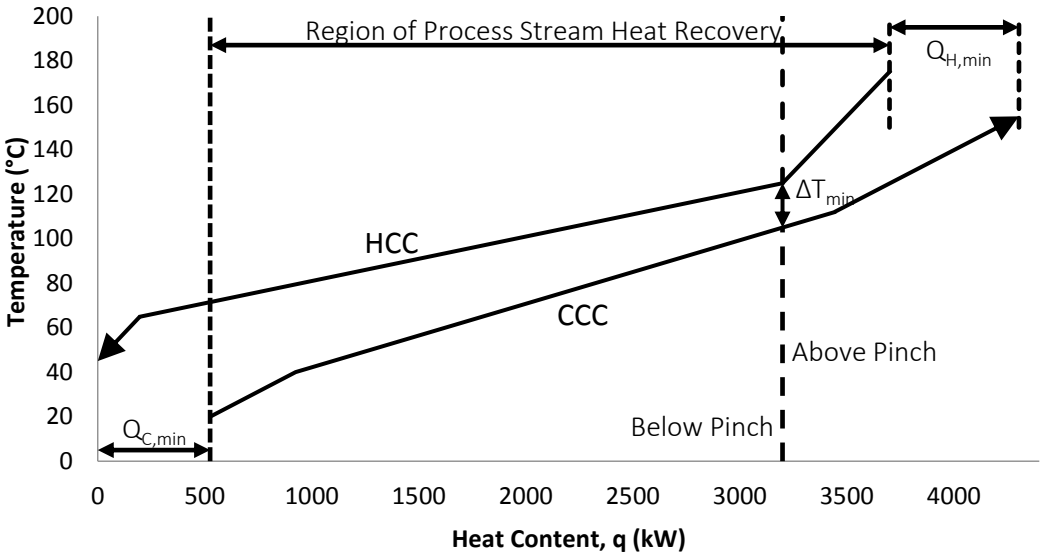

Figure 2.1: *A representative example of composite curves used to determine the minimum utility target.*

In Figure 2.1 the two composite curves are shifted horizontally so that there is a maximum overlap of each curve with the other curve, while still maintain the ΔT_{min}. The horizontal region over which the two curves overlap is where heat exchangers can be placed between process streams, whereas places of overshoot at either end of the diagram represent the minimum utility requirements for the hot and cold utilities. The point at which the composite curves are the closest vertically is the ΔT_{min} (or EMAT) and is known as the pinch point. This pinch point thus represents the bottleneck to energy recovery within the process and divides the problem into two independent regions that cannot be linked thermodynamically according to the PT method.

It can be noted that the optimal network may not be the one with minimal utility usage, nor the one that uses the particular ΔT_{min} used to set up the composite curves. Even with the options for stream matching greatly decreased in the process of energy targeting here, there can still exist many potential network designs that satisfy these energy target requirements but with different capital costs. The optimal HEN is therefore the one that minimises both the capital costs and the utility requirements, or total annual costs (TAC). The TAC can be divided into operating costs and capital costs. Operating costs are usually only represented by utility costs but should also include the pumping costs associated with the pressure drops as a result of the exchangers. While the capital costs are made up by a number of factors including the number of heat exchangers required, the total area of the heat exchangers, number of shell passes per exchanger, the materials of construction, and exchanger type. PT

has only concentrated on using the targeting concept on the minimisation of the number of units and the exchanger areas, with the other aspects of the design only accounted for after the network is developed.

2.2.2 Capital Cost Targeting

2.2.2.1 Minimum Units Targeting

One of the key determiners in the cost of a network can be the number of units, and a minimum number of units can be targeted prior to the network design. In each section above and below the pinch it is possible to express the minimum number of units by a formula based on Euler's Network Theorem shown below in Equation 2.1 (Linnhoff, et al., 1982):

$$N_{min\ units} = N_{streams} - 1 \qquad (2.1)$$

Where $N_{min\ units}$ is the minimum number of units in the section above or below the pinch point in question and $N_{streams}$ is the number of streams. This equation can then be applied to each region separated by pinch points. Before the development of area targeting, targeting using the minimum number of units was the most common method used to determine the optimal capital costs of the networks, however the size of each exchanger should also be included as it is a key determiner of overall costs.

2.2.2.2 Overall Exchanger Area Targeting

Area targeting came about as a result of the realisation that the minimum areas above and below the pinch points for a network can be predicted from the composite curve plots through the formulation of a balanced composite curve (BCC). The BCC is essentially the same as the composite curves of Figure 2.1, except that the hot and cold utilities are now included on the graph at the respective ends, resulting in the balanced hot composite curve (BHCC) and the balanced cold composite curve (BCCC). The BCC can then be divided into vertical enthalpy intervals at each point of inflection, represented graphically using dotted lines in Figure 2.2, below.

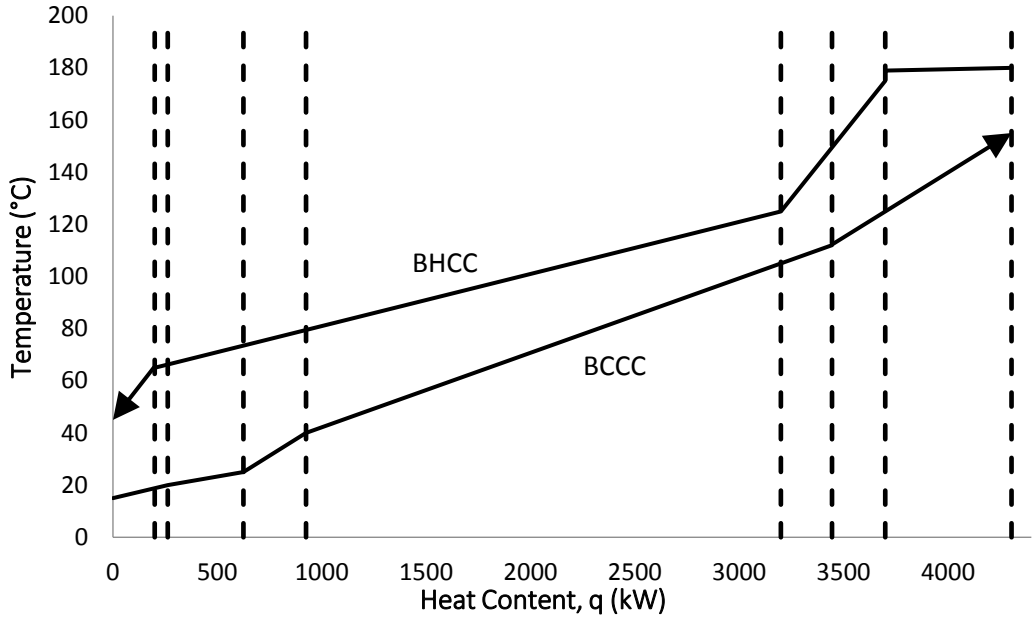

***Figure 2.2**: A representative example of balanced composite curves used in the determination of the minimum area target. Adapted from Shenoy (1995).*

In the above diagram "vertical heat transfer" would require that heat exchangers be placed within each of these intervals, using the temperature driving forces as shown on the diagram. Since the pinch is the area where heat transfer is most constrained, the units placed around the pinch tend to be large expensive units. Each of these intervals designated by the dotted lines in Figure 2.2 is treated as if it were an ideal counter-current heat exchanger with the area (A) calculated using Equation 2.2:

$$A = \frac{q}{U.LMTD} \qquad (2.2)$$

Where q is the quantity of enthalpy exchanged in the imaginary ideal exchanger, U is the overall heat transfer coefficient, and the logarithmic mean temperature difference is represented by $LMTD$ and calculated using Equation 2.3:

$$LMTD = \frac{\Delta T_2 - \Delta T_1}{ln\left(\frac{\Delta T_2}{\Delta T_1}\right)} \qquad (2.3)$$

Where ΔT_1 and ΔT_2 are the differences between the inlet temperature of the hot and outlet of the cold, and the inlet of the cold and outlet of the hot respectively. The LMTD is typically large (thus making the area small) when the temperature differences on either of the ends of the exchanger are large, providing greater driving force for heat transfer.

By summing all of these area, the minimum total area for the network (A_{min}) can be predicted. It is important to also realise that this procedure will only lead to a minimum area if the heat transfer coefficients of all process streams and utilities are equal (Nishimura, 1980). Linnhoff and Ahmad (1990) do note, however, that if the heat transfer coefficients for the exchangers do not differ by more than an order of magnitude then this area does not differ by more than 10 % to the actual area.

Assuming constant U for all exchangers is often a poor representation of reality as the individual heat transfer coefficients, h, are dependent on the stream properties and fluid velocities within each exchanger.

$$\frac{1}{U} = \frac{1}{h_i} + \frac{1}{h_j} \qquad (2.4)$$

Where i represents the hot stream and j represents the cold stream. By using Equation 2.4, Equation 2.2 can be re-written as the Uniform BATH Formula (Townsend & Linnhoff, 1984):

$$A_{network} = \sum_k \left[\frac{1}{LMTD_k} \left[\sum_i \left(\frac{q_i}{h_i} \right) + \sum_j \left(\frac{q_j}{h_j} \right) \right] \right] \qquad (2.5)$$

These values for h are typically derived using values obtained empirically or from "experience", and can also be derived using heat transfer correlations. Since these correlations rely on the fluid velocities, viscosities, and heat transfer data of the specific streams at the specific temperatures pertinent to each match, these are often estimated and assumed constant during the network synthesis with the details of the individual exchanger design only considered at the end of the network design. In addition, it is normal for a scaling factor to be associated with each exchanger as a thin film typically develops inside of the tubes of heat exchangers, forming a barrier to heat exchange that gradually decreases the performance of the exchanger over time. While these are subtle points, it should be kept in mind that these changes to the process parameters can have significant ramifications to the optimal solution once the design obtained is implemented.

Targeting of individual aspects of the network such as minimising utilities, number of units, exchanger areas, and shells, in turn can produce better networks than non-systematised approaches that were utilised before the invention of PT, however in order to find networks that have the optimal costs, some form of cost targeting is necessary.

2.2.2.3 Capital Cost Estimation and Targeting

The capital costs of a network that contribute to the TAC is a combination of both fixed costs, associated with the number of exchangers and shells, as well as of the area of each heat exchanger. Typically the installed cost of a single heat exchanger, $CC_{per\ unit}$, is represented by the following equation:

$$CC_{per\ unit} = a + bA^c \qquad (2.6)$$

Where a, b, and c are cost coefficients that are dependent on the type, materials of construction, and pressure rating of the exchanger.

Since it is possible to determine the minimum area of the entire network prior to the design from the Uniform BATH equation (Equation 2.5), as well as the minimum number of heat exchangers, U_{min}, it is possible to combine these to derive a capital cost target ($CC_{network}$) if a few simplifying assumptions are allowed. These assumptions are that the cost coefficients are identical for all exchangers, and that the areas of the exchangers are uniformly distributed (Smith, 2005).

$$CC_{network} = U_{min}\left[a + b\left({}^{A_{network}}/_{U_{min}}\right)^c\right] \qquad (2.7)$$

Where $A_{network}$ comes from Equation 2.5. These capital costs can then be annualised to attain the annualised capital costs (ACC) which can be added to the annual utility costs to obtain a TAC.

Attempts have been made to further refine this approach to account for a few of the limiting assumptions made during the cost targeting. Hall, et al. (1990) used weighting factors to each individual stream to allow for the considerations of exchangers that had non-uniform specifications, known as the "ϕ-factor" method. The method finds a single reference cost law, usually the one that can be applied to the majority of streams, and then adjusts the heat transfer coefficients of the individual streams that use cost laws other than the reference streams to "cost weight" them.

$$h_j^* = \phi_j h_j \qquad (2.8)$$

Where

$$\phi_j = \left(\frac{b1}{b2}\right)^{\left(\frac{1}{c1}\right)}\left(\frac{A_{min}}{U_{min}}\right)^{\left(1-\frac{c2}{c1}\right)} \qquad (2.9)$$

And $b1$, $c1$ are cost law constants for the reference stream, $b2$, $c2$ are cost law constants for the exchanger with the specific construction requirements, and h_j^* is the new modified individual heat transfer coefficient to be used in the calculation of the individual exchanger areas. This method has a number of weaknesses, as it cannot be applied when both streams to be matched require special exchanger types, and the area targeting is needed to be done at least twice to determine the associated cost targets. Additionally, as noted by Polley and Jegede (1992), the h-value of a stream is dependent not only on the individual stream, but also on the stream that it is matched with, the material of construction, and whether the fluid is shell-side or tube-side in the particular match.

Jegede and Polley (1992) attempted to build on this work by developing a targeting technique that targets the area and number of units between exchanger specifications, by making adjustments to the h-value that are match-specific. Their method also made use of a basic supertargeting method for the optimisation of the exchanger minimum approach temperature (EMAT).

2.2.3 Supertargeting

All of the targeting methods mentioned above are carried out at a set value of ΔT_{min}, often based on the experience of the designer. If a low initial ΔT_{min} is provided then the composite curves are allowed to be placed closer together vertically. This will often result in lower utility usage as the overlap between the CCC and HCC in Figure 2.1 can be significantly less. However, as the value for ΔT_{min} is reduced, the area required for the exchangers around the pinch is greatly increased. This is due to the fact that the driving force for the heat transfer is significantly reduced. This suggests that there is some optimal value for ΔT_{min} that exists which balances the gains of reducing the utility costs with those of capital cost investment as a result of larger exchangers. This problem can be further complicated when a large number of streams with multiple pinch points are considered, especially if the heat transfer characteristics of the individual streams differ drastically. Supertargeting attempts to find the optimal ΔT_{min} and is done by finding the TAC of a network for different values of the ΔT_{min} and plotting this on a graph to determine the optimal value of the ΔT_{min} (Shenoy, 1995).

In the PT method once the targeting is completed then the network can be designed that attempts to get as close to the targets set as possible.

2.2.4 Network Design

When the targets for the HEN design are set, the final design can selected by the designer through the assignment of units so that the targets are met as closely as possible. For vertical heat transfer, diagram (a) on the left in Figure 2.3 is observed, with hot stream A matching with cold stream C and hot stream B matching with cold stream D. If there are a large number of hot streams and a small number of cold streams, a large amount of stream splitting may be required in order for the vertical heat transfer requirements to be met. Networks with a large amount of splits are known as "spaghetti" designs (Smith, 2005).

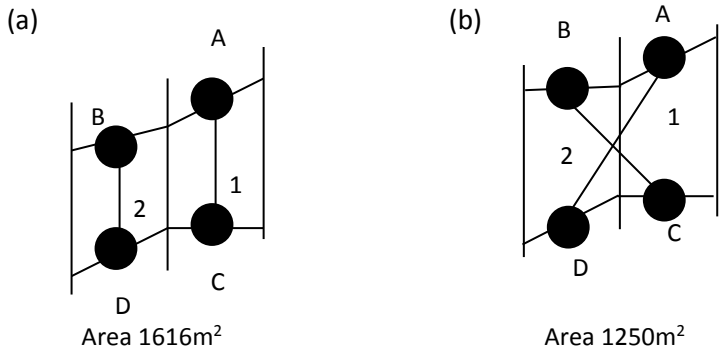

Figure 2.3: *Non-vertical (criss-crossing) heat transfer matching (Smith, 2005)*

For problems in which the individual heat transfer coefficients are significantly different, it may be necessary to relax the use of vertical heat transfer in order to meet the area targets that were set. In Figure 2.3 diagram (b), where the verticality concept is relaxed, hot stream A has a low film heat transfer coefficient and cold stream C has a high film heat transfer coefficient. In interval 2, hot stream B has a high film heat transfer coefficient and cold stream D has a low film heat transfer coefficient. Matching the streams with vertical heat transfer in (a) gives a total area of 1616 m^2, whereas matching with what is called "criss-crossing" gives a better area of 1250 m^2. The BC match, which uses lower temperature driving force, but where both heat transfer coefficients are high, uses this lower driving force more effectively, thus providing a smaller overall area for the network. The low heat transfer coefficients of the AD match require more driving force to produce a smaller exchanger due to the low heat transfer coefficients. While this is a useful analysis, it can be shown that individual heat transfer coefficients are not always the best method to estimate the overall

heat transfer coefficient, as flowrates of the individual streams within the exchanger greatly affects the overall heat transfer coefficient for each match (Smith, 2005).

There are a number of rules that can be applied in determining the optimal matches for a given network, which were determined by Linnhoff, et al. (1982). The rules of thumb, while not providing a rigorous guaranteed optimum, do lay the groundwork for practical and near-optimal networks that approach the pinch targets. The design approach does not, however, provide for orders of preference and ranking of importance to any particular matches, leaving the designer to decide on the best network. The rules are as follows:

1. No heat should be transferred across the pinch.

2. No cold utilities should be used below the pinch or hot utilities above the pinch.

3. Above the pinch the number of cold streams should be higher than or equal to the number of hot streams, and below the pinch the opposite should be true. If this does not hold then stream splitting cannot be avoided.

4. Since the pinch is the thermodynamic bottleneck in the problem, moving away from the pinch therefore provides better temperature driving forces. Matches should therefore be chosen in which the heat capacity flowrates of the cold streams are greater than the heat capacity flowrates of the hot streams above the pinch, with the opposite true for below the pinch. This will ensure that the temperature driving forces are used more efficiently.

5. The driving force plot (DFP) is a convenient way to determine whether the driving forces in individual exchangers is being utilised efficiently. The DFP is constructed by taking the temperature difference between the two composite curves and plotting this against the actual temperature of either the hot or cold streams. This diagram can be used to guide the design toward the minimum area target and analyse which design are over the target. In some cases the DFP will provide information on whether the duty should be reduced on certain exchangers in order for the driving forces to be used more efficiently, however this may lead to more units than the minimum (Serth, 2007).

6. Once the design is obtained, it may be necessary to rigorously calculate each area in order to make a quantitative assessment of the areas, and how the adjustments from the DFP may affect them. This takes a lot of effort if done manually, but it is possible to implement this step in computer software to optimise the continuous variables involved once the matches are set (Serth, 2007).

While these heuristics can be used to guide the design towards the targets set, the sequential manner of the approach can result in suboptimal solutions.

2.2.5 Limitations of the Approach

It should be noted that in all of these targeting methods a number of decisions are made by the designer during the design process that can trap the final design into a solution that may end up being sub-optimal. This is because the problem is decomposed into a number of sub-tasks that are performed sequentially. If the designer recognises this then a complete redesign is required. The disadvantages of these approaches are numerous. Even if the designer has access to extremely detailed costing models, the actual costs are not considered at any point during the design phase; only calculated at the end, with the areas or number of units acting as a proxy for the actual costs during the design. Additionally, the fixing of ΔT_{min} and the energy targets at different points during the design phase results in a trial-and-error approach. Another serious disadvantage in the sequential method is that stream matches across the pinch points cannot be considered. By excluding these matches many potential, unintuitive solution networks can be excluded. Furthermore, many detailed considerations of actual heat exchanger design are ignored during these procedures that can drastically affect the cost and viability of a generated network, such as the number of shells, pressure drops, the effect of changes in the heat transfer coefficients etc. Solutions that involve splitting streams are also difficult to consider, and thus these sequential design approaches cannot guarantee truly optimal networks.

Newer techniques have proven many of these statements true and that a simultaneous approach based on advances in computational power, solver technology and mathematical programming can provide better solutions. Section 2.4 goes into the details of these

methods that are quickly becoming commonplace in industry, however for the next section parallels between the HENS and MENS are drawn and the techniques developed that link these two separate fields are discussed, with particular focus on how PT has been applied to MENS.

2.3 Pinch Technology on MENS

The MENS problem was first defined in a paper by El-Halwagi and Manousiouthakis (1989), although the core concept had been a feature of process synthesis for many years before then. It is a more specific aspect of mass integration, whereby a systematic approach is developed that attempts to form a fundamental understanding of the flows of mass within a process system, utilising this understanding to identify and optimise the flows of species throughout a process (El-Halwagi, 1997). Mass exchange networks (MENs) are necessary in a diverse range of applications including feed preparation, waste treatment and reduction, as well as product recovery; taking the form of stripping, leaching, adsorption, absorption, extraction, and ion exchange. Mass exchangers are vital in pollution prevention and, in the terms of this thesis, are regarded as units that selectively transfer undesirable species (contaminants) from a waste stream, source, or rich stream, to a mass separating agent (MSA) via direct contact. MSAs are identified and chosen based on the contaminants and process fluids and need to be immiscible with the process streams, usually being contacted in a counter-current manner, similar to many heat exchanger configurations. In water networks, water is the only MSA, and, since the most and largest water-using operations are in fact not mass transfer operations, this large body of literature is not reviewed in this literature review (Alva-Argáez, et al., 1999).

MSAs, or lean streams, can be described as either once-through or regenerable. Once-through MSAs are used when the effluent MSA has become valuable in some way (as a feedstock or product) through the acquisition of the contaminant or if there is no economic or environmental benefit to regenerate the lean stream leaving the process. Regenerable MSAs provide overall economic or environmental benefit from having the solute recovered or from regenerating the MSA for recycle purposes.

Similar to HENs, MENs can involve a large number of streams with an optimal network difficult to determine because the minimisation of the TAC is made up of a trade-off between operating costs (made up of MSAs in MENs and utilities in HENs) as well as the exchanger capital costs. The exchanger sizes (heights and diameters of columns in the case of MENs and areas and shells in the case of HENs) are also a direct result of the flowrates of the respective streams and the driving forces that determine the rate of mass and heat transfer respectively.

For these reasons developments that have been applied to HENS have, to a large extent, also been transferred into the field of MENS. While there are many analogues between heat and mass transfer, Szitkai, et al. (2006) identifies 4 key differences that add additional complexities into mass transfer problems that need to be taken into account:

1. Equilibrium relationships between rich and lean streams need to be considered in order to calculate driving forces and mass exchanger unit sizes.
2. No analogy exists between HENS and MENS for the case of multiple contaminants.
3. In MENS the lean stream flowrates need to be determined, however in HENS the cold stream flowrates are given a priori.
4. There is no need for external rich streams in MENS, whereas in HENS, hot utilities are required in order for cold streams to reach target temperatures

Once all of the similarities and differences between the MENS and HENS problem were identified the application of HENS technology, which was already fairly developed, could be used to solve MENS problems.

2.3.1 Minimum MSA Targets

In the MENS-defining 1989 paper by El-Halwagi and Manousiouthakis (1989) they extended the pinch approach for HENS to the application of MENS by first targeting the minimum cost of MSAs without a predetermined network structure and then, in a second stage, minimizing the fixed cost by generating a network with a minimum number of exchangers. They presented a mass exchange composite curve using “corresponding composition scales”.

In this approach an exchanger minimum approach composition (EMAC) is defined, similar to the ΔT_{min} in HENS, which is used to ensure that the mass transfer is feasible throughout the

network. The EMAC differs to the ΔT_{min} in that it has to be defined separately for each stream, and is used in the corresponding composition scales in conjunction with the equilibrium relationship between the rich and lean streams to generate a one-to-one correspondence for streams that have thermodynamically feasible mass transfer. Using this formulation it is possible to represent all MSAs and rich streams on a single mass pinch plot. Figure 2.4, below is an example of this equivalent plot.

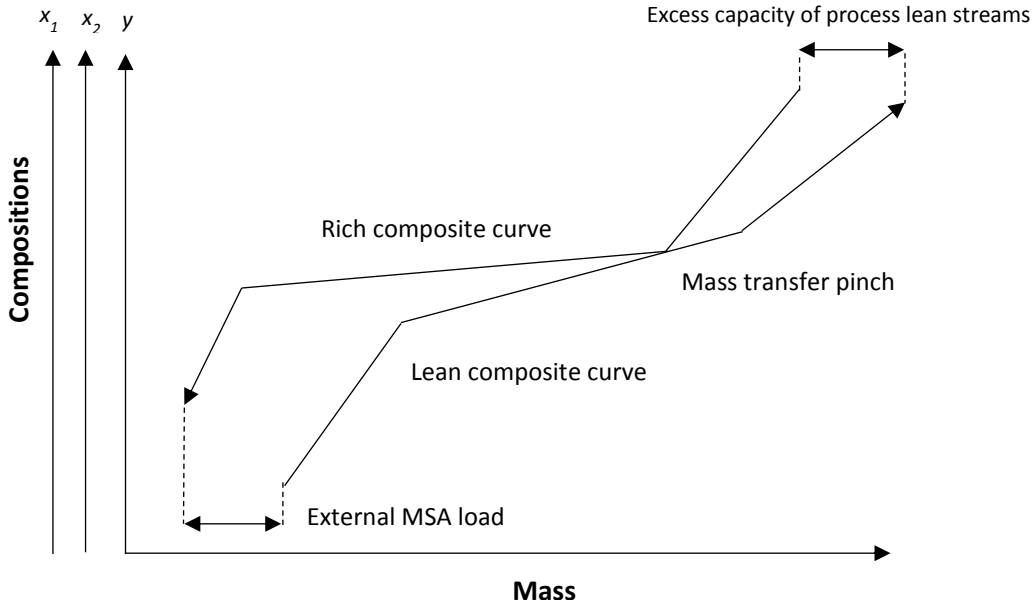

***Figure 2.4**: A representative example of a mass exchange pinch diagram. Adapted from El-Halwagi (1997).*

Figure 2.4 is very similar to the HENs pinch diagram, Figure 2.1, with a few key differences, namely that the pinch point here is where the rich composite curve (RCC) touches the lean composite curve (LCC). This is due to the fact that the EMAC is already included into the LCC using the corresponding composition scales approach discussed above. This point graphically displays the thermodynamic bottleneck of the system, in the same way as the pinch in HENS. In other ways these two diagrams are very similar, with the vertical areas of overlap representing the region where the maximum process to process mass exchange can take place, with no mass exchange possible across the pinch. The regions of overshoot at the horizontal extremities of the graph represent the excess capacity of process lean streams, and the need for external MSAs, similar to the requirements of cold and hot utilities in the HEN systems.

In the study of El-Halwagi and Manousiouthakis (1989), where this approach was developed, they continued by first targeting the minimum cost of MSAs without a predetermined

network structure, and then, in a second stage, minimizing the fixed cost of the network by generating a network with the minimum number of exchangers. In subsequent papers, El-Halwagi and colleagues (El-Halwagi & Manousiouthakis, 1990a, b, & c) made use of mathematical programming techniques to automate the pinch process and made use of more advanced targeting methods that included regeneration of the MSAs and non-isothermal operation. These techniques are reviewed in Section 2.4.3.1.

2.3.2 Capital Cost Targets

In a trend similar to the developmental history of HENS, the next developmental milestone in MENS was the inclusion of capital cost targeting. The number of exchangers in the network, heights, diameters, column internals, and materials of construction, all contribute to the TAC of the network in the form of the ACC.

The first method of establishing a target for capital costing was done in the same way as the first attempts associated with HENS, namely the act of targeting the minimum number of units. El-Halwagi and Manousiouthakis (1989) used the same formulation as was presented in Equation 2.1 for the HENS problem, applied below and above the pinch. With the large number of potential exchanger operations that can be considered, as well as the fact that the design of these operations can be complex, the other factors that contribute to the ACC of MENS were not considered for quite some time.

2.3.2.1 Mass Transfer Composite Curves

In heat exchangers, the capacity of the heat exchanger can be estimated predominantly from the area required to transfer heat, while the materials of construction, head type and heat exchanger type contribute only to the costing factors. In mass exchangers, the dominating factor for exchanger capacity can be assumed to be the height, with the internals estimated based on this height. Two of the most commonly used mass exchanger operations are trayed columns (stage-wise exchangers) and continuous contact exchangers (packed columns) and Hallale and Fraser (2000a & b) developed targeting techniques that attempt to find a way to target the sizes of these types of mass exchange operations before the network design is fixed.

The first contribution from the work of Hallale and Fraser (2000a) was the development of a modification to the mass exchange pinch diagram (shown in Figure 2.5). The modification made use of a "*y*-*y** composite curve" which is constructed by using the MSA composition in equilibrium with the rich stream (*y**), rather than the MSA's composition (*x*). This developed curve is shown in Figure 2.5, below.

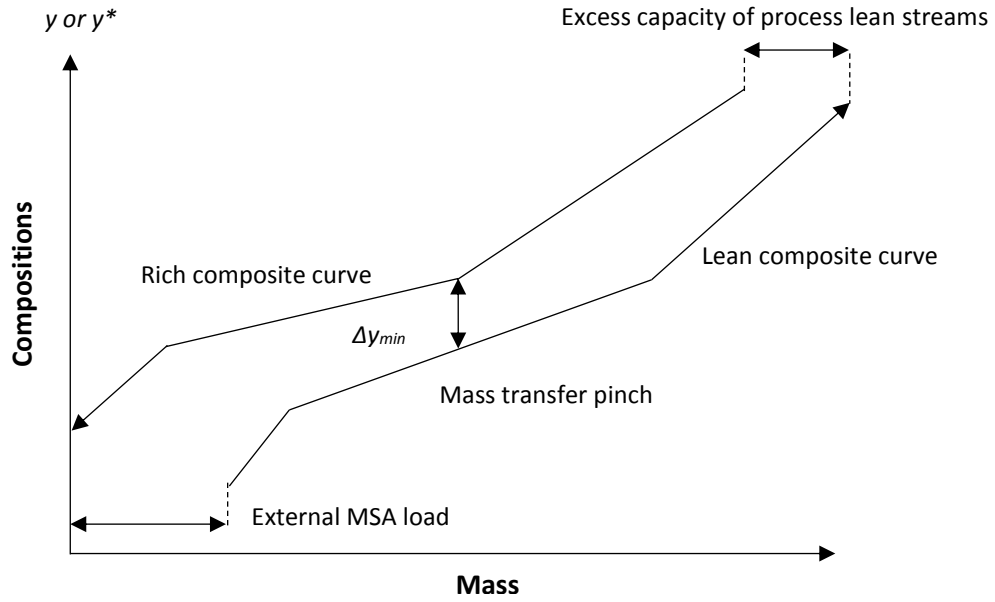

Figure 2.5: *Mass transfer composite curves developed by Hallale and Fraser (2000a)*

This formulation is a better analogy with the original pinch curve for HENS (Figure 2.1) and can thus be easily transferred for many of the same applications, including the targeting for minimum packed height or number of stages above and below the pinch. The advantage here is that, as opposed to the mass transfer pinch curve (Figure 2.4), the composite curves allow for many different MSAs to be combined into a single lean composite curve, as the *y** represents the contaminant's composition in the MSA in equilibrium with the rich stream (composition *y*). On the diagram Δy_{min} is the EMAC, with a direct analogue to the ΔT_{min} in HENS PT, and the vertical distance between the two composite curves represents the mass transfer driving force. This Δy_{min} differs slightly from the EMAC used by El Halwagi and Manousiouthakis (1989) in that it includes the equilibrium relationship between the rich and lean stream. Figure 2.5 can be used to size mass exchangers in much the same way as can be done with PT in HENS. The composite curves can be treated in the same manner as for HENS by partitioning the curve into composition intervals and treating each interval as a fictitious exchanger. These imaginary exchangers can then form the basis for the height targeting.

2.3.2.2 Mass Exchange Height Targets

Targeting procedures for continuous contacting columns, such as packed columns, can have very similar formulations to those used in heat exchanger network synthesis to determine the distribution of height amongst the various stream matches above and below the pinch. Hallale and Fraser (2000b) made use of a formulation similar to that used in the determination of the size of heat exchangers in order to determine an estimate for the minimum height of the network (Equation 2.10):

$$H_{min} = \sum_{k}^{intervals} \frac{1}{\Delta y_{lm,k}} \left(\sum_{i}^{Rich\ streams} \frac{w_i}{ky.a.XSA_i} \right)_k \qquad (2.10)$$

Where w_i is the mass load of rich stream i in interval k, $\Delta y_{lm,k}$ is the logarithmic mean composition difference (LMCD), ky is the overall mass transfer coefficient, a is the interfacial area of the packing, and XSA_i is the cross sectional surface area of the column. The ky, a, and XSA_i, can be estimated from the process parameters using flowrates, stream properties, and packing properties. Using Equation 2.10 in combination with the minimum number of units can help to distribute the number of units among the stream matches and intervals. It can be noted that during this procedure a large number of process parameters are estimated. The task of targeting is to identify the minimum height possible in the network. Since the height is determined directly from many of these estimated parameters, any changes to these estimates as a result of a detailed design of the individual units can drastically alter these estimates, and thus the entire procedure. This point is of great importance to this thesis as a whole.

2.3.2.3 Mass Exchange Stage Targets

For columns that involve stages, the number of stages per fictitious mass exchanger can be obtained through a graphical approach or via the Kremser equation if the equilibrium line is straight. Hallale and Fraser (2000a) point out that many operations often have straight equilibrium lines, however if curved equilibrium lines are required, only the graphical method can be used. The Kremser equation is shown in Equation 2.11 below:

$$N_{real} = \frac{\ln\left[\left(\frac{y_{i,in} - m_j x_{j,in} - b_j}{y_{i,out} - m_j x_{j,in} - b_j}\right)\left(1 - \frac{1}{A}\right)\right]}{-\ln(1 + E_M(A-1))} \qquad (2.11)$$

Where m_j is the slope of the equilibrium line, y is the rich stream composition, and x is the lean stream composition. A, known as the removal or absorption factor, is determined with Equation 2.12:

$$A = \frac{L_j}{m_j G_i} \qquad (2.12)$$

Where L_j is the flowrate of the liquid stream (absorbent or lean stream) and G_i is the flowrate of the gaseous stream (rich phase). E_M is known as the Murphree efficiency, which is used to determine the actual number of stages when unideal operation is assumed (i.e. equilibrium is not reached in each stage). It is calculated with:

$$E_M = \frac{y_n - y_{n+1}}{y_n^* - y_{n+1}} \qquad (2.13)$$

Where y_n^* is the lean phase composition that would be in equilibrium with the exiting rich phase. Once the minimum number of stages for each section above and below the pinch is determined, the stages can be divided among the minimum number of units to determine the targets for each section. In order to get an estimate for the costs associated with these columns, the stage heights and diameters need to be determined. Hallale and Fraser (2000a) made use of a 15 % inactive height in the column and used a correlation to determine the tray spacing based on the column diameter. The column diameter was also estimated using an empirical correlation which relies on the tray spacing and the superficial velocity of the gas. They therefore conclude that this step requires initial guesses and successive trial-and-error steps.

As can be seen from the costing correlations to be shown later, the costs are obtained from the height, which rely predominantly on the diameters. The number of trays plays a part in the determination of the cost, however height plays the most important role. When successive trial-and-error stages are required in order to obtain good targets, the targets lose efficacy. If the target is not the true minimum value, then it is not possible to design a globally optimal network based on these targets. The fact that the heights and diameters are based on loose approximations and a sequential calculation strategy means that it may be difficult to design a network that is globally optimal, and that once the network is actually found the solution may not be reproducible because the approximations may grossly under- or overestimate the actual sizes of the exchangers.

2.3.3 Supertargeting

Once these methods are used to determine the estimates for the capital costs of the network based on the initial pinch diagram, supertargeting can commence. The supertargeting for MENS is the same as with HENS where the EMAC is altered and a new pinch diagram is obtained. This is used to again determine the minimum number of stages or minimum height above and below the pinch. The TAC of this new network is calculated. This is repeated and tested over a large range of EMAC values until a plot of TAC vs EMAC is obtained in order to find the optimal value for EMAC and thus the best network (Hallale & Fraser, 2000a).

2.3.4 Network Design

In a manner similar to HENS, once the targets have been set a network can now be designed by selecting which rich streams will be matched with which lean streams. In his book on mass recovery network synthesis, El-Halwagi (1997) provides a set of rules for the design of MENs that closely match the targets set. Since the pinch remains the most constrained section of the network, the matching should begin about the pinch, with no mass transferring across it. The rules are as follows:

1. In a similar rule to HENS network design, the number of rich streams above the pinch must be less than or equal to the number of lean streams, with the opposite true below the pinch. Stream splitting will be necessary if this inequality does not hold true.

2. Above the pinch the gradient of the operating line should be greater than or equal to the gradient of the equilibrium line. The opposite should hold below the pinch. Stream splitting may also be required here in order to meet this requirement.

3. Similarly to HENS, a DFP can be made that plots the vertical distance between the composite curves versus the compositions of either the rich or lean streams. In doing this it is possible to ascertain whether the selected matches make the best use of the driving forces available, or whether certain exchangers require adjustment in order to make better use of the available driving forces (Hallale & Fraser, 2000a).

2.3.5 Limitations of the Approach

Many of the same limitations that apply to PT's application to HENS also apply to its application to MENS, as discussed in Section 2.2.5. The sequential manner in which the network is derived leads to suboptimal solutions, and the approximations and assumptions required to simplify the network formulation can result in poor estimates for the targets themselves. Incorrect targets will result in the exclusion of potentially optimal solutions. In addition, the resulting networks, once designed in full using detailed design equations that rely on thermodynamic input, can be very far from the approximated solution derived during the targeting phase. In MENs, as with HENs, the velocities of the fluids directly affect the overall mass transfer coefficients. In HENs the capital costing equations are mostly reliant on the areas of the individual exchangers, however in MENs the costs are related to the height, diameter, and the internals of each column. In pinch technology, as presented by Hallale and Fraser (1998) and Hallale and Fraser (2000a & b), only the packed height or number of stages has been targeted, with the internals and diameters largely being estimated or ignored. With this, key parameters in the search for truly optimal networks are ignored. In order to find the optimal networks it is possible to search the entire solution space simultaneously without sequential targeting steps. In order to understand these simultaneous approaches, it is first necessary to gain an understanding in optimisation techniques and their application to mathematical programming.

2.4 Mathematical Programming

Traditionally sequential (or hierarchical) strategies have been used to optimise chemical systems by making use of heuristics and chemical engineering knowledge. The PT approaches discussed in Sections 2.2 and 2.3 are examples of such approaches. These approaches, however cannot simultaneously account for all the interactions present within complex networks and also make use of numerous rules-of-thumb assumptions to simplify the problem. With advances in computing, simultaneous (or algorithmic) approaches using mathematical programming have gained much attention and have been shown to create networks that are significantly cheaper (Kravanja, et al., 2012).

This section of the literature review will focus on simultaneous optimisation techniques in which the goal is to describe the physical system using mathematical models, leaving the designs of the actual units, flowrates, temperatures etc. as variables that the selected solver environment attempts to find the optimal values for. The optimal solution in this context is with reference to an objective function that is usually to maximise the profitability or to minimise the TAC. This thesis makes use of available solvers, but does not add any significant contributions to the mathematical optimisation solver field. For this reason this section will be used to describe the methods used, and also to point out the ways in which these solvers fall short in a number of aspects, and how these shortcomings form one of the main motivating factors for this work.

In this section of the literature review the techniques that have been developed for optimising networks of units using superstructure-based approaches will be reviewed first, followed by a look at deterministic techniques developed for solving MINLPs, with a particular focus on the application to process flowsheet synthesis. Finally, the techniques that have been developed to optimise HENs and MENs, based on these simultaneous strategies, are discussed.

2.4.1 Mixed-integer Nonlinear Programming

Superstructures are commonly utilised in the representation of optimisation models of process systems as they make it possible for many candidate solution structures to be embedded within the model. These candidate structures, or topologies, are then assessed by a solver in order to find a solution that meets the set criteria of the constraints and objective function. The solution structure selected by the solver is indicated by the existence of the process option through the use of a binary variable. In addition to this discrete choice of process units, there also exist continuous variables that represent component flowrates, unit sizes, temperatures, pressures etc. The presence of continuous variables as well as integer variables means that these problems are modelled as MINLPs (mixed-integer nonlinear programs).

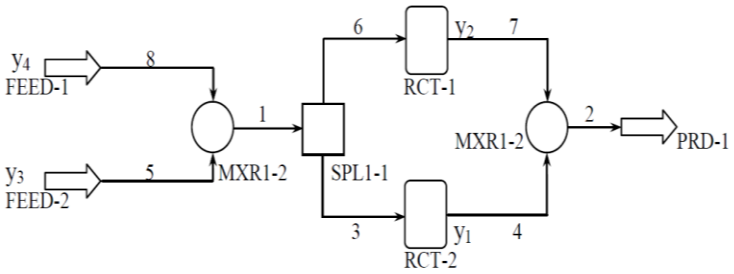

***Figure 2.6**: Representative example of a superstructure where a choice exists between two feeds and two reactor types (Kravanja & Grossmann, 1994).*

In the representative example, shown in Figure 2.6, the superstructure has embedded within it the choice between two possible feeds of varying compositions, FEED-1 and FEED-2, and two reactor types, RCT-1 and RCT-2, that are both possible options to create the desired product, PRD-1. These choices are represented by y_4 and y_3, and y_2 and y_1, respectively. The binary variables, y, therefore represent the structural options available to the model, whereas continuous variables are used to optimise the flowrates, conversions, reactor sizes, temperatures, etc. that are to be used. Mixers, MXR1-2, and splitters, SPL1-1, are also important units in these representations, as the presence of these often result in the presence of bilinear equations, which add to the non-convexity of the formulations.

MINLPs take the form:

$$z(y^K) = \min_x \; c^T y^K + f(x)$$

subject to:

$$g(x) \leq 0$$

$$h(x) = 0$$

$$A\,x = a$$

$$B\,y^K + C\,x \;\leq d$$

$$x \,\epsilon\, X = \{x | x \,\epsilon\, R^n, x_L \leq x \leq x_U\}$$

$$y^K \,\epsilon\, Y = \{y^K | y^K \,\epsilon\, \{0,1\}^m, Ey^K \leq e\} \qquad (2.14)$$

In the above representation (Equation set 2.14) x represents continuous variables like flowrates, temperatures, and vessel sizes, which are typically bounded by upper and lower values. The binary variables, y^K, represent the existence of process units at the *Kth* iteration of the program. $f(x)$, $g(x)$ and $h(x)$ are all nonlinear equations. In the objective function $z(y^K)$, $c^T y^K$ represents fixed costs and $f(x)$ represents the costs that are dependent on unit sizes and process requirements. Process specifications are included in the inequality constraints $g(x)$. Material and energy balances as well as design equations are satisfied using the equality constraints $h(x)$ and $A\,x = a$. Logical constraints, $B\,y^K + C\,x \;\leq d$ and $Ey^K \leq e$, are used to ensure that the selected flowsheet is within the superstructure (Kocis & Grossmann, 1987).

Yeomans and Grossmann (2000) found that MINLP models are able to yield superior solutions for chemical engineering flowsheeting problems compared to other methods, such as disjunctive programming, because they are able to embed a far greater number of possible flowsheets within them. A significant drawback of MINLP modelling, however, is that whenever a unit is excluded from the superstructure, all variables associated with such units tend toward zero. This can often result in discontinuities or singularities and thus the problem needs to be formulated carefully to avoid this potential pitfall. Additionally, the MINLP formulation also results in many redundant constraints that need to be solved at every iteration of the algorithm, resulting in greatly increased computation times (Yeomans & Grossmann, 2000).

In order to solve problems that are formulated as MINLPs, a global optimisation technique needs to be employed. Options for global optimisation are either deterministic or stochastic in nature. Stochastic techniques, such as genetic algorithms, Montecarlo minimisation, or

simulated annealing, do not make assumptions about the problem functions and make use of random elements in their search procedures, with the probability of a global optimum solution approaching 1 as solution time increases to infinity (Ryoo & Sahinidis, 1995). These solution strategies require high computing power and have very long computation times, and even when a solution is reached, it is never possible to determine whether a global optimum has actually been achieved.

Deterministic approaches, on the other hand, take advantage of mathematical structures inherent in specific problems and can often guarantee finite convergence within a certain level of accuracy, and are therefore more appropriate for flowsheet optimisation (Ryoo & Sahinidis, 1995). For this reason, it is important to formulate the MINLP in such a way that the specific solver being used can best exploit the mathematical structure of the problem to enhance the chances of a globally optimal solution being reached and also to speed up the rate of convergence to a solution. An understanding of the different deterministic solver methods is thus also required.

2.4.1.1 The Branch and Bound

The branch and bound strategy (Beale, 1977) implemented as SBB in GAMS, is the solver strategy with the longest history for mixed-integer problems. The strategy is an extension of the mixed-integer linear programming (MILP) solution procedure, which has a very well-studied resolution strategy and the ability to solve very large problems to proven global optimality. In this strategy a search tree is established where, at every node, a non-linear program (NLP) is solved for partial binary assignments. It can be thought of as a more brute-force approach as it exhaustively searches each “branch” of the “tree” of binary variables, however it is improved by saving the upper and lower bounds of the minimum solution and using these bounds in order to eliminate the “branches” of the solution space in which the optimal solution will not appear.

The method is successful in generating feasible solutions on extremely large problems that are poorly formulated, however the method is limited by the ability of the NLP solver itself, as are all MINLP solution strategies, as there is no known method to rigorously determine the optimal solution of NLPs in which there are non-convex terms. In chemical engineering applications non-convex terms are very common with bilinear terms almost always exist

around splitters and mixers, and capital costing and thermodynamic functions often being highly nonlinear.

The SBB solver will therefore still find trouble in finding global optima, and yet will often give solutions with extremely long computational times, which is especially undesirable in problems with a large number of binary variables.

2.4.1.2 Generalised Bender's Decomposition

The Generalised Bender's Decomposition (GBD) (Benders, 1962) strategy uses alternate solutions of NLP problems and MILP problems to be solved. This solution strategy is now commonly employed in a variety of MINLP solvers. In the GBD strategy, binary variables are fixed and the resulting NLP subproblem is solved as a continuous optimisation problem. This NLP solution then provides an upper bound to the objective function. An MILP is then solved in the next step, with the solution providing a lower bound, as well as new binary variables. The MILP master problem contains only the discrete variables and the Lagrangean cut in the binary space of the inequality constraints (Diwekar, et al., 1992). The new configuration is then solved as an NLP in the proceeding iteration. If the upper bound is less than the lower bound, then the new structure is the globally optimal solution, otherwise this procedure is repeated.

The advantage of this strategy is that special structures in the NLP subproblems can be easily exploited, however it often requires a high number of major iterations between NLP subproblems and MILP master problems (Viswanathan & Grossmann, 1990). This is because the MILP provides only loose bounds to the original MINLP problem as a result of the fact that the master problem MILP does not contain continuous variables (Duran & Grossmann, 1986). In order to improve on this, the outer-approximation solution strategy was proposed.

2.4.1.3 Outer-approximation

The outer-approximation (OA) (Duran & Grossmann, 1986) is similar to GBD in that it solves a sequence of NLP and MILP subproblems that provide the upper and lower bounds to the objective function respectively. The key addition to GBD that OA introduces is that the MILP master problem is formulated using continuous variables, as opposed to only integer variables in GBD. In the OA methodology outer-approximations are found at the solution

point of the NLP subproblem in order to construct a master problem that is made up of linearisations of non-linear constraints and the objective function.

By using the linearisations of the continuous variables for the MILP master problem it is possible to overestimate the continuous feasible region and underestimate the objective function. Since the master problem retains the continuous variables as well as the integer ones, the OA method requires the solution of a larger master problem than GBD. Although this could potentially add difficulty to obtaining the solution when many iterations are required, there is evidence to suggest that OA normally converges in fewer iterations compared to GBD (Diaz & Bandoni, 1996; Duran & Grossmann, 1986).

A large drawback of both the GBD and OA method is that certain convexity conditions are required to be satisfied so that convergence to a global optimum can be guaranteed (Diwekar, et al., 1992).

2.4.1.4 Outer-Approximation/Equality Relaxation

Kocis and Grossmann (1987) expanded the OA method so that it was capable of handling nonlinear equality constraints. The outer-approximation/equality relaxation (OA/ER) also makes use of NLP and MILP subproblems. However the method is able to add linearisations of the equations from the NLP subproblem to the MILP master problem, by utilising the signs of the Lagrangean multipliers at the solution to the NLP subproblem to relax them (Diwekar, et al., 1992).

The OA/ER algorithm requires strict convexity requirements if a globally optimal solution is to be found, with functions *g* and *f* assumed convex and the nonlinear equality constraints, *h*, assumed to be quasi-convex. If these conditions are not met, it is likely that the NLP subproblem will be trapped in local optima, as opposed to the globally optimal solution. Viswanathan and Grossmann (1990) thus proposed a further improvement.

2.4.1.5 Outer-Approximation/Equality Relaxation/Augmented Penalty

The Outer-Approximation/Equality Relaxation/Augmented Penalty (OA/ER/AP) attempts to improve the robustness of the OA/ER approach through more efficient handling of the non-convexities (Viswanathan & Grossmann, 1990). In this solution strategy an augmented penalty function is included in the MILP master problem. The solution strategy first solves

the relaxed NLP and then the master problem is formulated to include the slack variables. Violations of the linearisations of non-convex constraints are allowed in order to find feasible solutions to the master problem through the use of these slack variables in combination with the penalty function. By doing this the algorithm no longer requires the explicit verification of nonconvexities. This method has been shown to increase the robustness of solutions (Kravanja & Grossmann, 1994)

In an attempt to improve the solution generation times, as well as solve large highly non-linear systems and make use of the special structures that are inherent in process synthesis problems, a variety of techniques have been developed that use the OA and its variants to synthesise optimal process flowsheets. As can be seen, these techniques still rely heavily on the NLP optimisation for accurate optimal solutions to the NLP subproblem, in addition to reliable methods at incorporating more detailed information to accurately determine upper and lower bounds for the MILP master problem to get good estimates for the subsequent binary variables.

Because of this, the formulation of the process synthesis problem is extremely important. As will be seen in the subsequent sections, as well as in Chapter 4, 5, and 6, a lot of attention has been given to the formulation of MINLP problems that contain as many linear relationships as possible, with a minimum of bilinear terms, to as far as possible avoid non-convexities. Even with all of these measures in place and the resulting necessary model simplifications, it is still not possible to reliably assure that the problems are solved to global optimality, as the presence of local optima is difficult to avoid in the presence of all of the non-convexities involved with the capital costing equations, as well as the high number of binary variables in many of the problems.

2.4.2 Mathematical Programming and HENS

Mathematical programming has been the predominant tool for HENS strategies since the large-scale proliferation of computers from the late 1980's. The research has been carried out through the application of both sequential approaches and simultaneous optimisation through the formulation of LPs, NLPs, MILPs, and MINLPs. These formulations define the heat exchange network problem as a set of constraints linked together by variables and parameters to represent mass and heat balances. This model is then solved using a

deterministic or stochastic solver strategy in order to optimise an objective function. Sequential approaches dominated the field until the first uses of superstructures in HENS appeared with the introduction of the stage-wise superstructure of Yee and Grossmann (1990). Most of the sequential approaches have revolved around the automation of pinch technology.

2.4.2.1 Sequential Mathematical Programming Approaches to HENS

The automation of PT was the first use of mathematical programming for HENS. Among the first applications of PT was the transhipment model of Papoulias and Grossmann (1983). Their approach involved the formulation of an LP model for energy targeting, based on the problem table algorithm of Linnhoff and Flower (1978). After the minimum energy target is obtained, based on a set value for ΔT_{min}, an MILP model is used to obtain the target for the number of units. In a further application of PT, Floudas, et al. (1986), developed MAGNETS, an interactive program which uses the model of Papoulias and Grossmann (1986) to perform the energy targeting and number of units targeting, followed by a corresponding NLP that minimizes the area costs based on the heat loads and configuration from the solution to the MILP. With the addition of an NLP subtask, non-convexities are introduced into the model as a result of the bilinear terms around the mixers. As was discussed in Section 2.4.1, this means that a globally optimal solution cannot be guaranteed, with only local optimality assured. This method also has many of the same disadvantages associated with the pinch method in that sequential targets are set, without a truly simultaneous trade-off between all the costs associated with the network achieved.

The work of Papoulias and Grossmann (1983) was used as the basis for the work of Colberg and Morari (1990) as well. Their approach made use of different NLP formulations in order to optimise for the minimum area and capital costs at a specified heat recovery. This formulation allowed for streams with unequal heat transfer coefficients, different capital cost functions, as well as the ability to perform retrofit HENS. Jegede and Polley (1992) subsequently presented a modification to Colberg and Morari's model (1992) that allowed for the selection of non-uniform exchangers with heuristics and detailed cost functions used during the cost targeting phase.

Gorji-Bandpy, et al. (2011) used a multiple stage optimisation strategy that uses a genetic algorithm (GA) for the optimisation of the network topology optimisation and sequential

quadratic programming (SQP) in order to optimise the thermal load of the exchangers. Once the SQP obtains a solution for the optimal thermal loads the GA then uses these values to determine the fitness of the network. This 2-stage process was shown to improve the solutions obtained by standard simultaneous optimisations as well as pinch methods, however the use of stochastic methods means that the optimisation can be slow for large problems and there is no way to validate whether the solutions are globally optimal.

Serna-González and Ponce-Ortega (2011) used a novel method based on targeting methods to synthesis for an optimal HEN design. The problem was solved as an NLP optimisation problem which determines the optimal pressure drops and heat transfer coefficients of the streams in order to meet area and utility targets that are predetermined. The structure of the network is generated using a spaghetti design, which assumes heat transfer takes place vertically between balanced cold and hot composite curves. Their model has the disadvantage that it uses approximations for the pressure drops that are not necessarily comparable with heat transfer software packages and that the networks are fixed based on a sequential method similar to that of Papoulias and Grossmann (1983). This has been shown to result in poorer solutions to that of the MINLP approach where matches are unrestrained.

The shortcomings of sequential methods have been stressed throughout the literature review thus far and need not be stressed further. With the availability of many powerful modern solvers in commercial software packages and the increases in computer hardware, truly simultaneous optimisation strategies have seen a large rise.

2.4.2.2 Simultaneous Mathematical Programming Approaches to HENS

Sequential strategies involve the breaking up of the synthesis task into a number of subtasks, each done in order to guide the design to a solution. Simultaneous optimisation allows for all of the factors that influence the TAC, i.e. operating costs, area costs, and fixed costs, to be simultaneously considered. In a simultaneous approach it is also possible to consider networks that have exchangers that exchange heat across the pinch and to have no set value for ΔT_{min}.

Floudas and Ciric (1989) were among the first to attempt such a formulation. Their formulation initially uses the minimum utilities targeting approach in order to identify the pinch locations at a set ΔT_{min}. A hyperstructure is then introduced that attempts to represent all potential matches and network configurations within it. This is modelled as an MINLP

with the integer variables representing the existence of certain matches and an objective function based on capital costs. The MINLP formulation is non-convex and a special extension of the GBD algorithm was used which decomposes the non-convex networks into convex subtasks to identify upper and lower bounds for the master problem. The hyperstructure approach allows for the possibility of selecting a number of matches that is more than the minimum, an improvement on the Floudas, et al. (1986) NLP formulation, however the fact that the TAC is not considered and that ΔT_{min} is fixed, means that the optimisation is not truly simultaneous because a level of supertargeting is required to find the optimal ΔT_{min}. In a subsequent paper, Ciric and Floudas (1990) allowed for the model to be applied to a pseudo-pinch problem to allow for heat to flow across the pinch.

Further improvements on this method were made by not decomposing the problem into a transhipment problem followed by an MINLP, but rather including the transhipment model of Papoulias and Grossmann (1983) into the MINLP (Ciric & Floudas, 1991). In this formulation the authors were able to account for both the operating costs and capital costs simultaneously using both the pinch method or pseudo-pinch method and account for the heat loads and matches simultaneously within the hyperstructure. This model, however was highly non-convex with non-convexities in both the objective function as well as the constraints and resulted in local optima and difficulty in obtaining feasible solutions for large problems (Floudas, 1995).

Arguably the most influential and important development for the simultaneous synthesis of HENS is the superstructure-based approach of Yee and Grossmann (1990) and the many subsequent derivatives by various authors. These superstructure-based approaches are of great importance to this thesis and will be covered in detail in Section 2.4.2.4. Other approaches that are stochastic in nature will be briefly reviewed first.

2.4.2.3 Stochastic Mathematical Programming Approaches to HENS

While this thesis attempts to use deterministic solvers in order to find the optimal networks for HENS and MENS, other authors have attempted to use stochastic algorithms. The reasons for this decision are explained in Section 2.4.1. These stochastic algorithms will only be briefly mentioned in this thesis.

Lewin (1998) used an evolutionary algorithm (EA) to synthesise for cost optimal HENs. The novel evolutionary algorithm used in the study was based on the GA and was used to

optimise an NLP formulation of the problem. The study was done at constant ΔT_{min} and many of the matches were chosen based on the observation that most optimal networks contain fewer exchangers with fewer stream splits. The large degree of non-linearity is easily handled by the EA, however it is not possible to determine whether the solutions found are globally optimal and the time taken to solve the larger problems is significant.

Yerramsetty and Murty (2007) also used an EA approach for HENS, however their method did away with the isothermal mixing assumption for split streams and does not fix the ΔT_{min}, as was the case with Lewin (1998). The modifications present in their model also made use of binary variables, as opposed to the NLP model of Lewin (1998). This is possible through a differential evolutionary method (DEM) in which the mutations are directed in the direction of the optimal solution. DEM is more likely than DE to find a globally optimal solution, however it is not possible to guarantee a global solution, even with relatively large solution times reported, especially for large problems.

Bagajewicz, et al. (1998) introduced a state space approach to mass and heat exchanger network design. The state space approach is a process synthesis philosophy developed by Bagajewicz and Manousiouthakis (1990, 1992), finding application in a large number of chemical synthesis tasks. The basic premise is that any system can be described by a set of inputs and a set of output variables, related by a set Input-Output (I-O) relations. A set of variables known as state variables can be used to determine the outputs based on the inputs and I-O relations. By reducing the HENS/MENS problems into two operators (processes where transfer takes place and processes where splitting or mixing takes place) it is possible represent the synthesis problem succinctly and use this formulation to determine optimal networks, generating similar topologies to both hyperstructures and superstructures. This approach also cannot guarantee globally optimal solutions and is solved as an NLP in much the same way as many other approaches.

This thesis aims to use deterministic solvers to find optimal solutions for these problems. These deterministic approaches can allow for rigorous determination of optimality conditions, as opposed to the stochastic methods which rely on structured guesswork in order to find a brute-force solution. These brute-force approaches can be applied to many different structures of problems, however they often take extremely long times to reach solutions, and there is no way to guarantee an optimal solution. The deterministic approaches, described in Section 2.4.1, rely very heavily on structured formulations in order

to take advantage of the various solver methods, as convexity and optimality conditions can be readily exploited with intelligent problem formulation.

2.4.2.4 Superstructure-based Mathematical Programming Approaches to HENS

The most commonly-used, extensively applied, and modified approach to HENS in a deterministic solver environment is the approach first posited by Yee and Grossmann (1990). In this formulation a superstructure is presented that allows for the problem to be partitioned into a structure that embeds as many possible network topologies as possible into it, and then formulating the remaining equations to allow for the solver to find the optimal network that exists within this structure. This is made possible through the selection of binary variables to represent the existence of exchangers between hot and cold streams, and continuous variables that represent the heat flows, intermediate temperatures, and flowrates. Throughout this thesis, there is a constant reference to various superstructure approaches and formulations, and many of the details found in the models are included in the relevant chapters in the literature reviews pertinent to each specific chapter. In this section of the literature review, therefore, much of the analysis will be brief, with the majority of the literature and model details presented in the relevant chapters.

Yee and Grossmann's (1990) superstructure formulation is the stage-wise superstructure, SWS, shown in Figure 2.7 below. Throughout the thesis this model will be referred to as the SYNHEAT or SWS model. In this figure the two hot streams and two cold streams pass each other in counter-current fashion, with temperatures decreasing from left to right. In each interval it is possible for each hot stream to match with each cold stream, including the option for either stream to split. Utilities are placed at the ends of the superstructure and are used to satisfy the final target temperatures of the respective streams. Figure 4.1, in Chapter 4, shows all possible matches in this example, however in this chapter only the superstructures will be presented. The number of stages, or intervals, is recommended to be set to be equal to the number that is larger between the number of cold streams and number of hot streams.

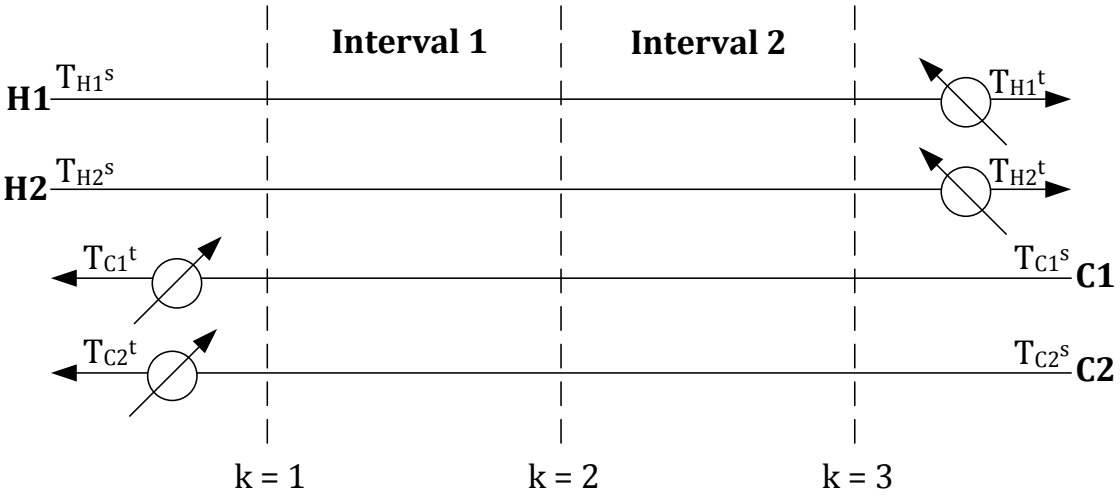

Figure 2.7: *Example of an SWS for two hot and two cold process streams.*

Optimisation of the Yee and Grossmann (1990) model involves the formulation of an MINLP. This MINLP, shown in detail in Appendix 4B of Chapter 4, is quite difficult to solve, especially as the number of streams increases. Since MINLPs are still a challenge to solve to global optimality, the model is formulated using approximations to represent the heat exchanger design in order to simplify the formulation. These approximations allow for linear constraints; particularly if isothermal mixing is assumed because this assumption can allow for the omission of a bilinear term in the mixing equation. To account for the possibility of isothermal mixing, the original formulation included a further NLP sub-optimisation, which fixed the exchanger topology and relaxed the isothermal mixing assumption. While having all constraints linear for the MINLP, the formulation still has a large non-linear, non-convex objective function. The assumptions made in order to reduce the size of the MINLP means that the superstructure omits the following structures, highlighted by Floudas (1995):

- Splits with two or more exchangers in series on one branch
- Bypass streams that are only feasible for non-isothermal mixing
- Any combinations involving these two structures

Despite these drawbacks, the model is still used in a large number of modern approaches and has been shown to provide good HEN structures and been robust enough to handle large problems. The details of this formulation and its utilisation in this project are included in Chapter 4, however it needs to be pointed out at this stage that the use of linear constraints, while useful in solving the model to optimality, only provides a rough estimate of the final network. Once detailed designs of the individual heat exchangers within the network are taken into account, such as the effects of fluid velocities and dynamics on the

overall heat transfer coefficients, the design constraints related to standard heat exchanger design configurations, pressure drops, etc. the final network may look significantly different in terms of capital costs to the “optimal” solution found within the SWS formulation. While the heat and mass balances are not approximations, the resulting areas can be very far from a real design. In addition to this, all of these simplifications do not manage to guarantee that the solution found from the optimisation is a global optimum, only that it is locally optimal and special initialisation and bounding techniques are required for larger problems in order to find feasible solutions. This problem is evident throughout all of the methods used for network design throughout the literature, and this is a key issue that the method presented in thesis aims to rectify. Many of these problems are avoided in the method presented in Chapter 4.

Much of the research subsequent to the SYNHEAT model’s first implementation and into the mid-2000s attempted to find new ways to reformulate the constraints so that globally optimal solutions could be found more easily or could be guaranteed. This often came at the cost of model complexity. One of the first of these reformulations was that of Daichendt and Grossmann (1994) in which a preliminary screening procedure was implemented on the SYNHEAT model. The transhipment model of Papoulias and Grossmann (1983) was reformulated as an MILP in order to use this as an aggregated model to undertake screening of the potential solutions. This aggregated model is used as the upper bound for the number of units and is used to indicate where matches exist for the SYNHEAT model. In doing this the authors attempted to provide tighter bounds for the problem, as well as reducing the superstructure alternatives. This was hoped to reduce the computing times and allow for a higher chance of attaining globally optimal solutions.

Zamora and Grossmann (1998) used a different technique that involved a hybrid branch and bound/OA optimization algorithm. The model used new convex formulations for the heat exchanger areas that ensure that the objective function is underestimated, thus allowing for good initial bounds for the minimal TAC. Their model used simplifying assumptions of no stream splitting, linear cost functions, and an arithmetic mean temperature difference.

Bjork and Westerlund (2002) used the SYNHEAT model to find another method of ensuring global optima can be reached by reformulating the problem by convexifying signomial terms. This special reformulation also allows for the isothermal mixing assumption to be removed, however at the cost of less accurate solutions, with a piece-wise linearization being required.

The method also has the drawback of not being guaranteed to reach global optimality in large problems (Verheyen & Zhang, 2006).

As optimisation algorithms developed, and computation power increased while also becoming more accessible, the determined manner with which the optimisation community attempted to attain global optimality, despite the NP-hard nature of the problems, decreased. This led to new attempts at finding increasingly complex and detailed ways of obtaining optimal networks through novel superstructures and the addition of more detailed constraints to allow for more realistically represented networks to be considered.

The SWS approach involves the use of undefined intervals where all streams participate in each interval. Isafiade and Fraser (2008a) used a similar framework to the SWS, however used the supply and target temperatures of either hot or cold streams to define the interval boundaries. This approach, called the interval-based MINLP superstructure (IBMS), had two iterations, the cold-based IBMS and hot-based IBMS, in which the supply and target temperatures of either the cold or hot streams are used to define the interval boundaries respectively. For the hot-based IBMS all of the cold streams are available for heat transfer in every interval, whereas the reverse is true of the cold-based IBMS. The approach also made use of the utility streams to define intervals, allowing for multiple utilities to easily be incorporated. The method made use of the same model formulation as the SYNHEAT model. An example of the IBMS is shown in Figure 2.8, where in each interval hot and cold streams are allowed to split and match with each counter-current stream.

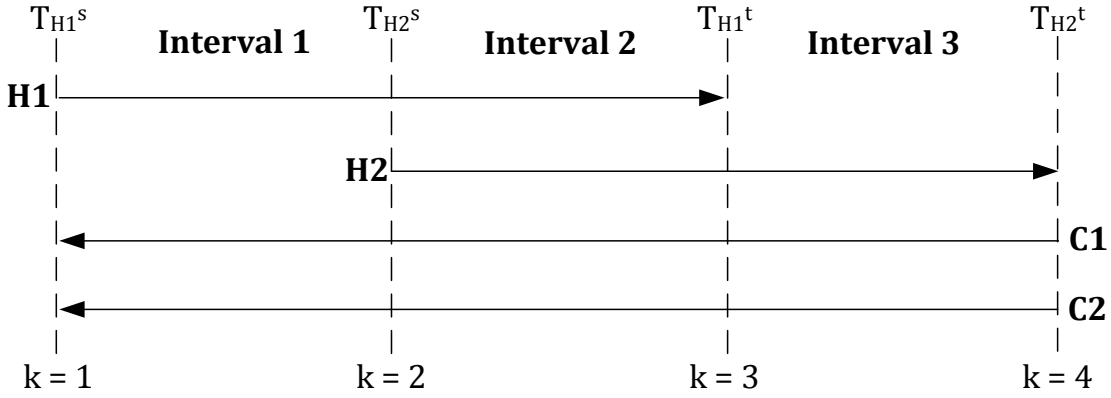

Figure 2.8: *Example of a hot-based IBMS for two hot and two cold process streams.*

This approach was applied to many examples and was proven to be more effective in some instances than the SWS. By defining the intervals in this way it is possible to firstly create more intervals than are present in the SWS, when using the recommended number of intervals from Yee and Grossmann (1990), thus allowing for more potential networks to be

generated. In addition, streams with vastly different temperatures are separated, thus reducing the number of extraneous binary variables that are unlikely to be used. By defining utilities in a similar manner to process streams and by allowing for them to define intervals also allows for more potential solutions that do not just rely on a similar approach to the pinch where utilities can only be considered to make up for a lack of process heat. Finally, the method reduces the need for initialisations and bounds, thus reducing the search region and allowing for higher chances of globally optimal solutions for the solvers.

Along similar lines, Azeez, et al. (2011), developed the target and supply based superstructure (T&SBS) and the supply and target based superstructures (S&TBS). These superstructures used both the hot and cold streams to define the intervals, with the T&SBS defined using the target temperatures of the hot streams and the supply temperatures of the cold streams and the S&TBS using the supply temperatures of the hot streams and target temperatures of the cold streams to define interval boundaries. An example of the S&TBS is illustrated in Figure 2.9.

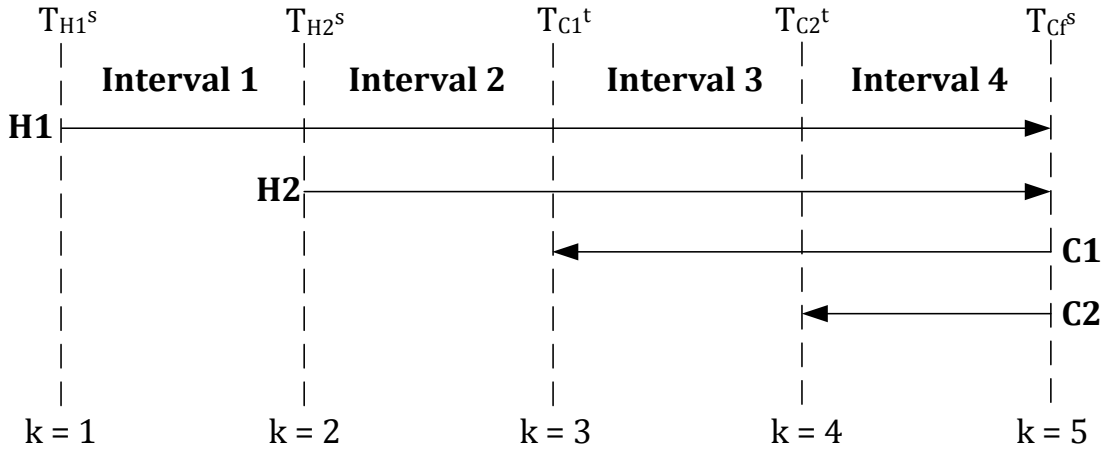

Figure 2.9: *Example of the S&TBS for two hot and two cold process streams as used by Azeez, et al. (2011).*

It is necessary in these two superstructures to define a fictitious temperature boundary. For the S&TBS this is the final temperature location, most probably a cold supply temperature that is likely to be the supply temperature of the cold utility. The opposite occurs for the T&SBS as the first temperature is a fictitious hot supply temperature, usually the hottest utility supply temperature.

Following these two proposed superstructures, the supply based superstructure (SBS), was subsequently introduced (Azeez, et al., 2012). This formulation made use of the supply

temperatures of both the hot and cold streams to define the intervals of the superstructure and is illustrated in Figure 2.10.

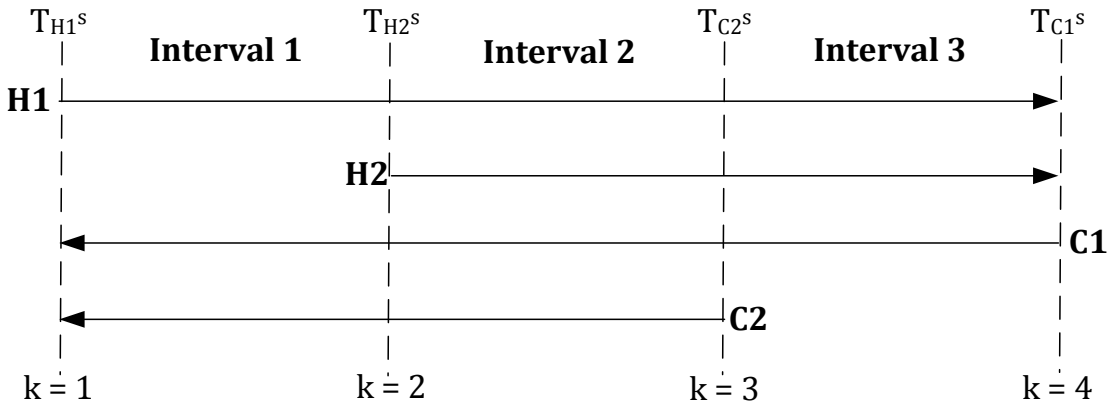

Figure 2.10: *Example of the SBS for two hot and two cold process streams as used by Azeez, et al. (2012)*

This model proved to be one of the most consistent performers of the three formulations as it uses the most extreme cold temperatures and most extreme hot temperatures, allowing for many potential matches along the superstructure. Azeez, et al. (2012) found that no particular model consistently finds the best solution however. The authors noted that increasing the number of intervals does not necessarily provide better solutions but models that use a higher percentage of the intervals created tend to perform better (Azeez, et al., 2012). These methods do not significantly alter the performance of the SWS, as the model formulations remain mostly intact, leaving the problems associated with the model still apparent; namely the use of approximations in the exchanger areas, isothermal mixing assumptions, and the non-convex objective function.

The other significant contribution to superstructures in HENS is that of Huang and Karimi (2013). In looking for alternatives to the superstructures of Floudas, et al. (1986) and Yee and Grossmann (1990), they presented 2 novel superstructures and model formulations. The first superstructure presented is a stage-wise, “match-centric”, approach that makes it possible for the model to select cross-flows, cyclic matching, matches in series on substreams within each stage, and multiple utilities that can be placed at any stage in the superstructure. This resulted in a formulation that was bloated with additional inequalities. Efficient deterministic solvers, like DICOPT cannot be used to solve the model, whereas the highly inefficient BARON solver is only able to find a solution with a maximum solution time set. While the superstructure approach presented by Huang and Karimi (2013) was shown to be able to select configurations that had hitherto not been considered, the extremely long

solution times and highly non-convex formulation means that the model is not going to be effective for very large problems and that global optimality is not assured.

The second superstructure presented by Huang and Karimi (2013) involves an "exchanger-centric" superstructure. In this superstructure there are no stages, rather only a number of exchangers that all the hot and cold streams can potentially make use of. The uniqueness of this approach is without doubt, however the authors failed to get competitive solutions due to trouble experienced in translating the superstructure into an efficient MINLP formulation. The formulation used had wider bounds on temperature variables, more binary variables, and has fewer nonlinear constraints, resulting in trouble in solving larger problems. Both of these formulations made use of the formulation of Huang, et al. (2012), explained below.

Huang, et al. (2012) improved the SYNHEAT model through the addition of non-isothermal mixing. The formulation adds in a number of constraints to effectively minimise the superstructures redundancies, thus reducing computation times. Through additional binary variables, stream bypasses are also allowed. Variables and constraints are also included that can be used to calculate the split streams' flowrates and to allow for non-isothermal mixing. These novel constraints are demonstrated in the paper to improve the efficiency and quality of the solutions obtained. Bjork and Westerlund (2002) used a similar approach, however their formulation did not also propose bounds on these newly included variables in order to improve the model's ability to deal with a variety of problems, which the Huang et al. (2012) formulation does to great effect. The authors also make use of numerous LMTD approximations in the paper to test their efficacy in a range of examples. While the authors do not believe an approximation is necessary, they show that the Paterson (1984) approximation is able to find the optimal network more often than the unapproximated LMTDs used in the study. The authors therefore propose a solution strategy that involves a subsequent solution involving the unapproximated LMTD (with constraints) after the initial solution and network configuration is obtained using the Paterson (1984) approximation. While these strategies are effective, the authors also point out the numerical difficulties that were still experienced. In the study DICOPT, one of the most efficient and widely used MINLP solvers, could not be used due to the non-convex nature of the problem and the lack of initial conditions. The authors explain that they were forced to use the less efficient and slower BARON solver with limits required to be set on the solution times due to the extremely slow convergence times that were experienced (up to 48 hours reported).

From these deterministic approaches it is obvious that the task of HENS is not trivial and the large non-convex problem cannot easily be solved to global optimality. Attempts to embed more potential networks within the superstructure by the addition of intervals, or by using non-isothermal mixing, rarely yield better solutions as the added complexity merely makes the problem more difficult to solve. In a study by Escobar and Trierweiler (2013) a variety of HENS approaches were compared with the suggestion that that systematic initialisation procedures for MINLP SYNHEAT formulations are required in order to increase the robustness of the approach. Their systematic comparison demonstrated, as had been previously thought, that the SYNHEAT model can generate higher quality networks than sequential strategies, with MINLP solvers DICOPT and SBB (discussed in section 2.4.1) the best and most consistently performing solvers. For these reasons the original SYNHEAT model is still used in the majority of applications of the simultaneous strategies. These issues with large models become even more evident when multi-period HENS is considered.

2.4.2.5 Multi-period HENS

Most HENS strategies have focused on optimisation of the network considering fixed parameters. However it is rarely the case in reality that heat exchangers operate at consistent temperatures and flowrates. Two possible dynamic situations are observed. The first results from uncertainties that occur when a particular parameter fluctuates around a nominal value, caused by poor control and/or changes in the environment. The ability for the HEN to deal with such changes is referred to as a network's flexibility. Periodic changes in operational conditions can also occur with changes in season or product specification. Heat exchangers that are designed to be optimal in multiple periods are referred to as multi-period networks (Verheyen & Zhang, 2006). In this thesis, the extension to multi-period operation is considered because the benefits of the proposed approach in HENS are especially obvious when considering this case. Since a large amount of the literature on multi-period HENS surveyed is included in Chapter 5, as well as the model details of the different approaches, this section will highlight the major contributions only.

The first to develop the concept of designing HENs capable of dealing with a wider range of processing conditions was that of Marselle, et al. (1982). They introduced the concept of resilience of a network. After defining a number of worst-case scenarios and manually designing an optimal network for each case, the best network is chosen based on the insights

of each case and the designers' judgement. The large number of cases makes the problem very large and the selection of the optimal network is not a simple or intuitive process.

Swaney and Grossmann (1985) used a flexibility index in order to determine key aspects about the flexibility of a design. The flexibility index represents the maximum deviation of uncertain parameters while still remaining within the feasible region and is useful for comparing the flexibility of a number of designs and for determining which areas of the design are limiting and thus the most critical in finding a flexible design.

Aaltola (2002) simultaneously optimised multi-period heat exchanger networks using a novel MINLP formulation which expanded Yee and Grossmann's (1990) SYNHEAT to multiple periods. Multiple periods are described beforehand with stream flowrates and the target and supply temperatures differing between each period. The formulation utilised the superstructure and most of the constraints from the SYNHEAT model, however also added a new set which was used to assign periods. The method also forms an MINLP first and this MINLP uses the average of the exchanger areas used over all the periods in the objective function. The exchanger capital costs are underestimated because of the use of the average exchanger areas over all the periods. An LP/NLP model and search algorithm is thus used after the network topology is generated in order to get an accurate TAC (Aaltola, 2002).

A model that deals with uncertainties in stream temperatures and flowrates was presented by Chen and Hung (2004). Their model broke the design process into three separate sequential stages. The procedure first used the SYNHEAT model to find a network with minimum TAC. This network was then tested for flexibility in all of the periods of operation and the networks that did not meet the flexibility criteria were removed through the use of integer cuts.

Verheyen and Zheng (2006) improved on Aaltola's (2002) model by using the maximum exchanger area instead of the average in the MINLP's objective function. This resulted in the addition of non-linear constraints to the model. An NLP suboptimisation is also added so as to include non-isothermal mixing. The formulation was shown to give superior networks in comparison to previous authors. The approach has proven to be the most commonly applied methodology for the MINLP synthesis of multi-period HENs and forms the basis for the MINLP step presented in this thesis. Chapter 5 elaborates on this method.

Chen and Hung (2007) made use of a multi-step procedure to design flexible mass exchanger networks and heat exchanger networks. The first step involved an MINLP optimisation

considering finite periods and then the networks are tested for flexibility by altering parameters. If the network passes the feasibility test for flexibility then the network is selected, if not the network is re-designed with a further MINLP optimisation to generate another candidate network (Chen & Hung, 2007).

Isafiade and Fraser (2010) extended their IBMS approach for single period heat exchangers to multi-periods and included the ability to optimise for periods that were not of equal duration. The model made use of the formulation of Verheyen and Zhang (2006) as its basis.

Ahmad, et al. (2012) used a simulated annealing algorithm on the model of Verheyen and Zhang (2006) to avoid problems with deterministic algorithms in finding global optima in non-convex solution spaces. The stochastic approach proved effective, however the expected longer computation times were experienced.

Jiang and Chang (2013) decomposed the multi-period problem of Verheyen and Zhang (2006) into a number of single period optimisations and then proposed a time-sharing plan in order to design more flexible heat exchanger networks that can be closer to optimality if the period lengths are not defined. The strategy was shown to be able to produce better TACs than previous networks, however cleaning schedules involved with sharing of process streams were not taken into account.

Sadeli and Chang (2012) analysed weaknesses in the MINLP formulations of multi-period HENS and developed a novel synthesis procedure, using an MINLP optimisation for the initial network, followed by a set of heuristics in order to include exchanger time-sharing. The initial network was generated in the same way as Verheyen and Zhang's (2006) model, however used F_T correction factors, calculated externally in MATLAB, once the MINLP was solved. These F_T correction factors were then used in an NLP subproblem where the network topology is fixed. Once the network is found the heuristics are used to find time-sharing schedules, with additional auxiliary heat exchangers included when needed.

Nemet, et al. (2012) used both stochastic and deterministic MINLP models in order to optimise a HEN over the entire lifetime. The authors identified the fluctuating prices of utilities over the lifetime of a plant and considered multiple forecasting scenarios for utility prices, using Net Present Value (NPV) as the objective function. The authors found significant differences in the solutions of the new models and those considering fixed utility costs, with the new models having significantly less utility consumption, resulting in increases in NPV.

Escobar, et al. (2014) proposed a heuristic algorithm that used Lagrangean decomposition to exploit the block diagonal structure of the multi-period HENS problem. This method postulates feasible solutions from Lagrangean decomposition subproblems in an iterative scheme and then uses these to update the multipliers with a subgradient method. Their decomposition strategy found similar or better solutions for all numerical examples evaluated, but found that the computational effort was only reduced for large problems. The heuristic approach found it possible to determine solutions from single period subproblems, meaning that the problems' solvability was dependent on the number of process streams, rather than the number of periods. While the method was limited by the fact that feasible solutions were generated heuristically, the optimality was shown to be guaranteed within a tolerance that lies within the dual gap (Escobar, et al., 2014).

Isafiade, et al. (2015) made use of Verheyen and Zhang's (2006) formulation and applied a binary variable reduction technique that involved the solution of multiple MINLP models in order to obtain different networks. These different networks were then used to limit the number of binary variables involved in a subsequent optimisation. The solutions obtained improved on previous studies for the examples considered and showed the effectiveness of reducing binary variables in order to reduce the size of the solution space in order to increase the speed and efficiency of the MINLP solvers.

Isafiade, et al. (2017) used an MINLP to integrate renewable energy options into multi-period HENS in a multi-objective optimisation strategy. The method made use of additional sets that included seasonal changes, as well as daily changes. This allowed for the model to include various renewable technology options for the generation of the utilities, taking into account the amount of renewable energy available during the day and in each season, as well as their respective costs and impacts to the environment. This greatly increased the size of the problem in terms of the number of binary variables, however the inclusion also ensured that a higher level of detail was incorporated than had been included in previous models.

It can be seen from the above studies that multi-period HENS has received significant study over the years, however much of the recent research has consisted of trying to find ways to decompose the problem into smaller problems. The formulations necessary to include accurate exchanger areas, as well as to account for a large array of operating conditions means the inclusion of a number of non-linearities and a great increase in the number of binary variables. These inclusions make the multi-period problem even more difficult to

solve, with the search for feasible solutions difficult in large problems and no guarantee of globally optimal solutions. Similar approaches have been applied to the MENS field, with problem formulation also proving to be difficult.

2.4.3 Mathematical Programming in MENS

The MENS problem has, unsurprisingly, largely followed the same trajectory as HENS. The first applications of mathematical programming techniques were applied to decomposing the synthesis problem into the subtasks involved in the pinch method and solving each sequentially to determine the optimal network.

2.4.3.1 Automated Sequential Methods for MENS

In an application of the procedure used by Papoulias and Grossmann (1983) to MENS, El-Halwagi and Manousiouthakis (1990a) made use of two targeting stages to determine the optimal network for single component systems. The first targeting stage made use of an LP formulation to locate the pinch and determine the minimum MSA usage. The second stage involved the solution of an MILP to determine the minimum number of units that meet the minimum MSA target. This procedure is performed at a set value of EMAC and ignores capital costing.

In a subsequent paper (El-Halwagi & Manousiouthakis, 1990b) the process was automated and regeneration introduced by dividing the set of lean streams into subsets of once-through and regenerable MSAs. In the paper the authors formulated the first stage as an MINLP with the objective of minimizing the lean stream and regenerating agents' costs, providing pinch points and optimal lean stream flows prior to the network design. The second step was formulated as an MILP in order to determine the network that satisfies these targets with the minimum number of transfer units (El-Halwagi & Manousiouthakis, 1990a, 1990b). El-Halwagi and Srinivas (1992) also used a similar principle to synthesize isothermal reactive MENs.

These procedures suffer the same weaknesses of pinch methods mentioned previously, with the additional weakness that only the number of units is considered; with the methods for

capital cost targeting in MENS being only established in 2000 by Hallale and Fraser, as previously explained in Section 2.3.2.

2.4.3.1 Simultaneous Mathematical Programming Approaches to MENS

Papalexandri, et al. (1994) presented a hyperstructure model analogous to the hyperstructure model for HENS of Ciric and Floudas (1989). Following this method, the hyperstructure attempted to embed all possible combinations of rich and lean stream matches and use this in an MINLP formulation for the determination of minimum TAC. The use of the Kremser equation (Equation 2.11) to determine the number of column stages, the mixer and splitter equations, and the splitting of the problem into a number of subnetworks that all contain binary variables, make this problem extremely non-convex with a number of bilinear terms. The model requires great computational effort and often produces sub-optimal solutions as a result. In addition, the costing is determined only by the number of column stages, a gross simplification of the intricacies involved in costing a MEN.

Comeaux (2000) simplified the model formulation of Papalexandri, et al. (1994), adopting pinch principles to reformulate the MENS problem as an NLP of moderate size. The superstructure approach employed ensured that each rich stream could potentially exchange mass with every lean stream, however it ultimately relied upon insights taken from a pinch approach and was shown to often result in suboptimal solutions. In a number of cases it gave solutions poorer than the fully simultaneous approach of Papalexandri, et al. (1994).

Chen and Hung (2005) used the superstructure approach of Yee and Grossmann (1990) and extended their model to handle multiple components to be removed from the rich streams, as well as to include non-isocompositional mixing. They formulated their model by using the maximum sized exchanger, based on all of the components that are transferred to the lean stream. The authors extended their approach to include reactive mass exchangers and problems with regeneration. Attaining globally optimal solutions proved to be problematic due to the non-differentiable max operator used in determining the number of trays required in each component exchanged as well as in deciding which height or number of trays was required to be added into the objective function.

The most significant contribution to simultaneous MENS arguably came from Szitkai, et al. (2006) who applied the SWS superstructure to the problem as well as incorporating the more

detailed capital costing formulations developed by Hallale and Fraser (2000a & b). Szitkai, et al. (2006) used a similar approach to Yee and Grossmann's (1990) approach to HENS in the synthesis of MENs. Their model was formulated as an MINLP and they presented a mostly linear single contaminant model as well as proposing a multiple contaminant model with the addition of nonlinear constraints. Their approach was best suited to single contaminant problems with packed columns as mass exchangers. They made use of the integer-infeasible path MINLP (IIP-MINLP) (Sorsak & Kravanja, 2002) model formulation to add stability and enhance the search for numerical solutions. Their model made the assumption that mixing streams must have the same compositions, limiting the solution space, but removing non-linearities. This is referred to as the isocompositional mixing assumption. This approach formed the basis for many other superstructure-based approaches and is presented in more detail in Chapter 6.

Emhamed, et al. (2007) introduced a hybrid approach to MENS that combined both Hallale and Fraser's (2000) supertargeting approach with the MINLP optimization strategy of Szitkai, et al. (2006). The method uses integer cuts and a bounding strategy in order to improve on the solutions generated in subsequent iterations and avoid infeasible networks. Emhamed, et al (2007) were the first to explicitly express diameter and packing costs into the objective function, however the diameters were fixed for each stream based on the flowrates and remained the same, regardless of whether the flowrate was split or not. The solutions from this method were also found to not be globally optimal.

Isafiade and Fraser (2008b) applied their IBMS superstructure to the MENS problem. The authors found that the new method provides different solutions to the model of Szitkai, et al. (2006), despite using the same model formulation, and in some cases providing better solutions; showing that their solutions are most likely locally optimal and underlining the problems associated with MINLP solvers. The authors suggested that, by defining stages in this way, it is possible that the problem is more efficiently initialized and bounded thus resulting in more efficient solution times. Isafiade and Fraser (2009) extended their IBMS approach to combined heat and mass exchange networks (CHAMENS) in a subsequent publication.

Azeez, et al. (2012, 2013) applied the S&TBS, T&SBS, and SBS superstructures to MENS. These new superstructure formulations were applied to a host of example problems and it was found that by re-formulating the superstructure in this way, new solutions could be found, however there was no conclusion as to which of the approaches provides consistently

good solutions. The methods were applied to both staged and packed columns. The SBS, by providing the highest chance of producing optimal solutions due to its large composition intervals, is explained in more detail in Chapter 6.

Liu, et al. (2013) used an NLP formulation to find optimal MENs with multiple components and used a genetic algorithm-simulated annealing algorithm hybrid approach (GA-SA). Their method does not rely on key components and uses number of trays to account for capital costing. They present only a small example that fails to find the best solution, even with a simplified superstructure in their formulation, again justifying the need for deterministic algorithms to efficiently solve larger problems.

Isafiade and Short (2016) made use of Azeez, et al.'s (2013) SBS formulation to determine cost-optimal MENs, with the addition of extra details for packed columns. These details allowed for the diameter to be included as a variable. With the inclusion of a variable diameter, the effect of velocity on the overall mass transfer coefficient, as well as the effects of flooding were also taken into account. This is done through the use of approximations detailed in Chapter 6 of this thesis. The isocompositional mixing assumption, present in most simultaneous MENS approaches was also removed. The additional constraints were highly nonlinear and required a special bounding and initialisation procedure in order to find feasible solutions. There is no way of guaranteeing globally optimal solutions, however these new inclusions showed the importance of taking into account detailed aspects of the individual exchanger's design at the network synthesis stage.

What can be seen from these modern approaches is not only that they follow the HENS approaches quite closely, but also that MENS has received less attention from the process synthesis community. In addition, the difficulties involved in the formulation of such problems are numerous, with relatively simple formulations proving challenging to solve with the current generation of MINLP solvers. Most papers presented here assume constant mass flowrates, and equilibrium conditions that do not rely on other components in the mixture, or the velocities of the fluids, as well as constant pressures and temperatures along the length of the column. The simplification that mass transfer can be approximated using the LMCD, and that the diameter can be assumed constant, even for streams that are split in most cases, are huge simplifications that will affect the final solutions and result in suboptimal solutions when the network is designed using more detailed analysis of the individual units. To understand the precise effect of the simplifications that are used in the

MINLP superstructure-based HENS and MENS formulation, it is important to consider the methods used and intricacies involved in designing the individual units involved.

2.5 Individual Unit Design

The shortcut or simplified models that are used in the network synthesis are necessary for a number of reasons that have been explained in preceding chapters. Firstly it is extremely difficult in sequential approaches for the designer to account for so many design factors and to predict the response of the system to each design change at one time. By decomposing the problem into multiple stages, it is possible to identify the main factors separately and then use this information to guide further designs. This is repeated for multiple iterations until a satisfactory design is obtained. In simultaneous approaches, it may be possible to formulate a problem in such a way so as to account for all design factors at once, however the combination of state-of-the-art mathematical techniques and modern computational power is still incapable of dealing with such large formulations. For this reason the process synthesis community has generally attempted to find ingenious ways to reformulate the problems in order to exploit the mathematical structures involved to be able to solve large problems. This often comes at the cost of losing accuracy. In either circumstance, network designs have been achieved by designing the network using approximations, shortcut models, and targets. Once the network topology is decided, the details of each individual exchanger have often been ignored in academia, or designed in industry, with little emphasis placed on how the actual detailed exchanger designs affect the optimality of the network design that was based upon the simplified exchanger representations. This is the gap that this thesis aims to explore and address. In order to do so, it is important to note the ways in which these units are designed, so that it is possible to understand how the intricacies of the design can affect the overall network solution.

2.5.1 Individual Heat Exchanger Design

There are a number of types of heat exchanger (HE): spiral, plate, plate-and-shell, plate fin, pillow plate, microchannel, double-pipe, shell-and-tube, direct contact, etc. Since shell-and-tube heat exchangers (S&THEs) are the most commonly utilized in industry and also the most studied, this is the only type of exchanger that is considered in this thesis. This section will only briefly delve into the methods used to design S&THEs, as the detailed method used in the case studies in this thesis is shown in explicit detail in Appendix 4C in Chapter 4. This section is included to provide for the reader that has little knowledge of HEs to be able to gain insight into the complexities involved in deriving accurate designs and how the inclusion of these can drastically alter the solution of the optimal network.

2.5.1.1 Challenges Associated with HE Design

Figure 2.11 shows the functioning of a 1-pass S&THE. In this exchanger the tube-side fluid enters the exchanger on the left and exits on the right, with the shell-side fluid entering on the right and exiting on the left. The tube side fluid is distributed among many small tubes on the inside of the exchanger in order to allow for as much of the fluid to be in contact with the heat transfer area, namely the metal tube surface. The shell-side fluid passes by a number of baffles, and therefore does not flow in direct counter-current flow to the tubes. This is a hindrance to heat transfer, however without the use of baffles it is likely that numerous dead-zones would appear within the exchanger in which little to no flow would occur, thereby not allowing heat transfer to occur effectively, and also resulting in the build-up of excessive scaling in these regions.

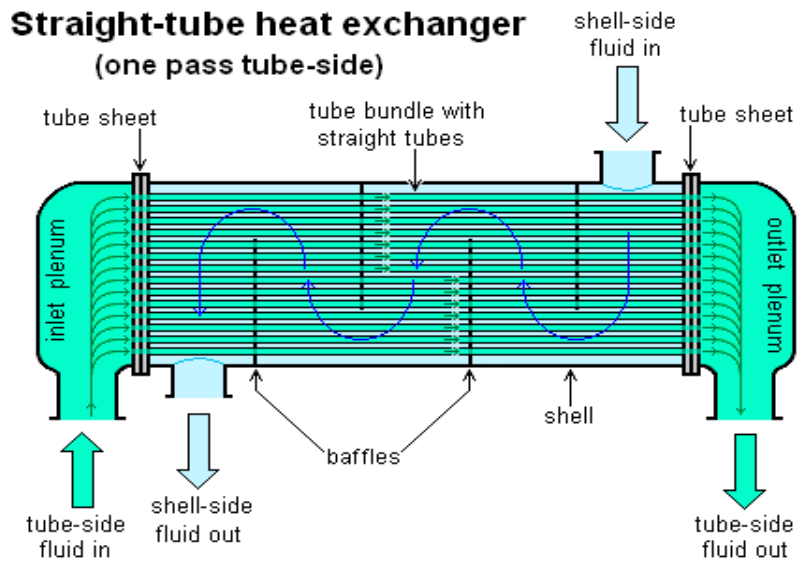

Figure 2.11: *Diagram representing a 1-pass shell and tube heat exchanger (Wikimedia Commons, 2016).*

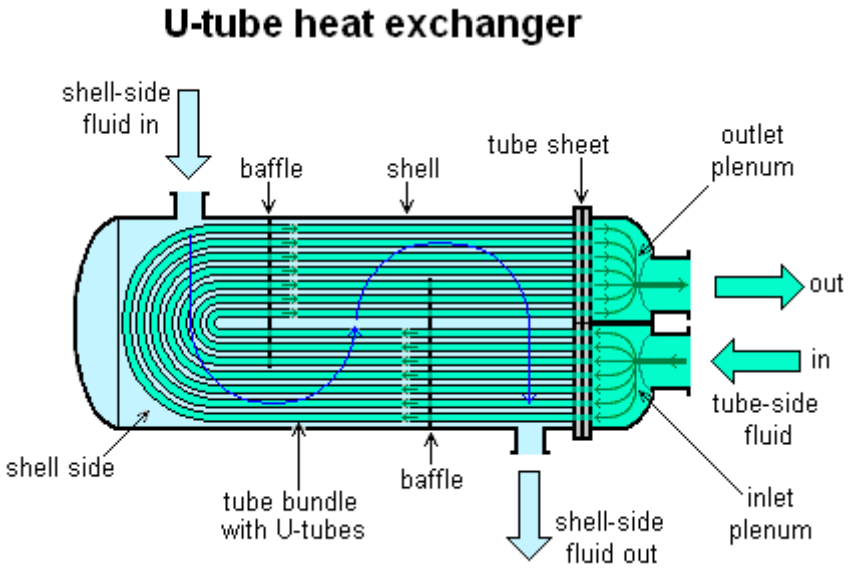

Figure 2.12: *Diagram representing a 2-pass U-tube shell and tube heat exchanger (Wikimedia commons, 2016)*

Figure 2.12 demonstrates what occurs in a system where we have two tube passes. In this design the tube-side fluid enters and exits on the same side of the exchanger, with the shell-side fluid again entering and exiting at opposite ends. This design means that the tube-side fluid's velocity is increased greatly, as the number of tubes is effectively halved, however

pressure drops are increased. In addition, counter-flow behaviour of the two fluids is now almost non-existent. This can mean that the assumption of counter-flow that is present in the network examples above will greatly underestimate the size required for the exchanger in question. To get an idea as to the degree to which non-optimal flow conditions can be realised, Figure 2.13 shows the complex flow patterns that are present for shell-side flow.

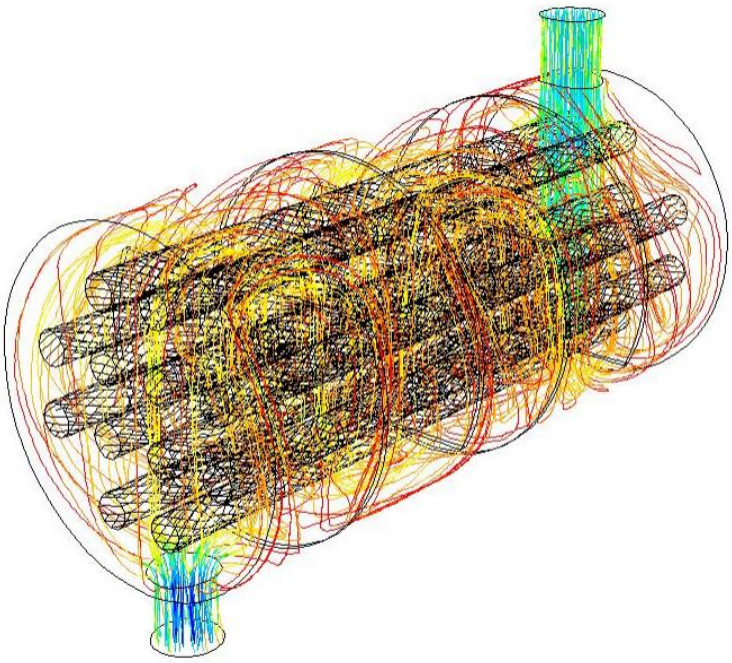

Figure 2.13: *Diagram showing the complexities present in a describing shell-side fluid flow in shell and tube heat exchangers (Wikimedia commons, 2016).*

The selection of a match between two fluids within a network could be suboptimal if design considerations such as those demonstrated in Figures 2.11, 2.12, and 2.13 are not included. Correction factors have been derived that attempt to correct for the suboptimal flow patterns for a wide variety of exchanger configurations. These are analytically derived and it is typical that the configuration is tweaked until this correction, known as the F_T correction factor, is less than 0.75, i.e, not increasing the ideal exchanger size by more than a factor of 1/0.75, or 1.333. This is a significant number and can be decreased by increasing the number of shells (Serth, 2007). The increase in the number of shells also has a significant impact on the final capital investment of a network and has not properly been dealt with in the network design stage of the methods available in the literature. This technique and its effects on designs is covered in detail in Chapter 4.

Of importance in HE design is the velocity of the fluids. By increasing the number of tubes, the heat exchange area is increased, while decreasing the velocity of the fluid. Decreasing the fluid's velocity has the negative effect of decreasing the heat transfer coefficient, as well as encouraging scaling. Scaling occurs from a build-up on the tubes as a result of minerals from the fluid leaching onto the tube, impairing heat transfer and increasing maintenance requirements. Decreasing the number of tubes has the opposite effect of increasing fluid flow while decreasing overall transfer area. High velocities can result in excessive mechanical stress on the exchanger, as well as excessive pressure drops. Furthermore, the selection of the number of tubes is further complicated by a range of factors.

When purchasing heat exchangers, there are a range of standard tube sizes and exchanger sizes, including the number of tubes and size of the shell, standardised by the Tubular Exchanger Manufacturers' Association (TEMA). This introduces discrete choices in the design procedure if the desired outcome is to design an industry-standard exchanger for use in operation. In addition, the choices for head types, tube layouts, baffle cuts, sizes, number of baffles, fluid allocation (tube-side or shell-side), and the effect of the velocities on the viscosities and heat transfer coefficients make the process of designing the optimal heat exchanger an extremely taxing task. It is little wonder that there has been little research into attempting to find the optimal HEN using detailed design information.

Typically the design of S&THEs has been done through a set of heuristics that help to guide the designer to an acceptable design, based on the experience of industry, empirical correlations, and an iterative procedure to test whether different designs are suitable for the task and this method is described in detail in Chapter 4 (Serth, 2007). There have been some attempts at optimising the complex system using mathematical programming, however.

2.5.1.2 Optimisation of Individual HEs

Mizutani, et al. (2003) made use of an MINLP reformulation of a GDP that optimizes shell-and-tube heat exchangers based on Bell-Delaware correlations to the shell-side flow and takes into account many detailed design variables including number of tubes, baffles, head types, internal and external tube diameters, and fluid allocation. The model minimizes a total annual cost that includes area and pumping costs. The model only considers single shells and

does not account for the TEMA considerations. It was found difficult to find the optimal solution in large problems.

A detailed MINLP model for the optimisation of individual shell-and-tube heat exchangers based on TEMA standards and including a detailed Bell-Delaware method was presented by Ravagnani and Caballero (2007). They modified the GDP model presented by Mizutani, et al. (2003) to include tube counting tables that involve many discrete decisions in order to optimise a heat exchanger according to TEMA standards. Due to the size and combinatorial nature of the problem, the authors found that using a branch and bound solver was more suited to the task than the more often-used decomposition algorithms. They rigorously bounded and initialised the problem and their models had 53 discrete variables and 713 continuous variables in 166 equations. With such a large number of binary variables the authors found it very difficult to find feasible solutions and therefore the solutions obtained were most likely to be locally optimal. The model improved on the model of Mizutani, et al. (2003) by including TEMA standards and also considering multiple tube and shell passes. The number shell passes did not modify the objective function, however, but did have an effect on the pressure drops. Unfortunately the authors did not make use of an example with multiple shells, so it was difficult to judge the effect of this on the solutions.

Vengateson (2010) used finite difference calculus in order to model the number of shells required for E and F type shells as well as to develop temperature profiles across all heat exchangers. The usefulness of the approach to individual exchanger design was also shown with capital cost equations used to compare solutions.

Sanaye and Hajabdollahi (2010) used a sorting genetic algorithm (NSGA-II) to optimise 2 objective functions and produce Pareto curves for individual heat exchangers considering maximum thermal effectiveness and minimum cost. The model used Bell-Delaware calculations for the shell-side and the tube arrangements, diameters, pitch ratio, length, number, and baffle spacing and cuts were considered as the seven design parameters. This approach took extremely long times to solve, with no guarantee that the solution was optimal.

Onishi, et al. (2013) used a sequential optimisation approach for the synthesis of near-optimal individual shell-and-tube heat exchangers by formulating a GDP problem as an MINLP optimisation. Their results show that decomposing the large highly nonlinear mixed-integer problem into smaller, well-thought-out objective functions can result in a very good

solution that takes into account many of the competing variables as well as using some of the insight of the designer.

Fettaka, et al. (2013) made use of a genetic algorithm and multi-objective optimisation to synthesise optimal individual exchangers with respect to area and pumping costs as individual objectives. The model makes use of detailed equations for individual exchangers and found that a single objective function may be insufficient to get an optimal solution as they can converge on solutions that are not globally optimal and are also very sensitive to the weighting chosen for the objectives in the single objective function.

It is possible to see from the above studies that it is difficult to find the optimal solution in HE design using deterministic solvers, mostly because of the large number of binary variables required if TEMA options are considered, as well as the binary variables required for baffle cuts, number of baffles, and fluid assignment. These binary variables, combined with the highly nonlinear equations that are required for the viscosity calculations at the specified velocities and temperatures, and for the F_T correction factors, make the task extremely taxing and none of the authors approached satisfying and reliable solutions using their methods. For this reason, it is still the industry standard to use a set of empirical correlations, combined with detailed heuristics and designers' experience in order to find optimal heat exchangers. One such method is described in detail in Chapter 4.

2.5.2 Individual Mass Exchanger Design

In the case of mass exchangers it is possible to create far more rigorous models than in the case of heat transfer as the fields of thermodynamics and mass transfer are quite developed, with column simulation research at a mature stage. The methodologies utilised in the optimisation of networks of mass exchangers, as has been showed in Section 2.3, Section 2.4.3, and shown in detail in Chapter 6, make use of the logarithmic mean concentration difference (LMCD). The LMCD makes use of a similar assumption as that of the LMTD in HENS, and is useful in determining the driving force for the mass transfer. Its application to MENS, in particular to trayed columns is not a very accurate representation of mass transfer, however. This in combination with the Kremser equation to determine the number of stages, which adds further discontinuities and nonlinearities, makes the formulation of the MENS MINLP model for staged columns challenging. In contrast, continuous contacting columns

allow for mass transfer to take place in an analogous way to heat transfer in HENS, with mass transfer taking place along the entire column, based on the composition differences along the column, as opposed to the discrete equilibrium stages of staged columns. In the case studies considered in Chapter 6, the predominant purpose of MENS is for the decontamination of gaseous process streams by absorption into liquid MSAs. For this purpose packed columns are often used as they are cheaper to operate because they do not necessarily require reboilers and condensers.

For these reasons the packed column has been studied in detail in this thesis, with the recommendation for further studies to attempt to include trayed operations. The shortcut methods that have been used in MENS up until now have been covered for both trayed columns and packed columns in Section 2.3.2. Since detailed analyses of the methods used in this thesis are provided in Chapter 6, this section aims to provide the reader with some background knowledge to better understand that chapter and its context in the process synthesis field, and will thus be brief in some of the more technical details.

2.5.2.1 Introduction to Mass Transfer

In mass transfer operations it is typical for at least two phases to be in contact. Since it can be assumed that the "bulk" concentrations within the gas and liquid are constant at each point along the length of the column, the mass transfer can be assumed to take place in a film that exists on each side of the interface between the two phases. In this film it is assumed that molecular diffusion takes place. Figure 2.14, on the next page, aims to illustrate this phenomena, known as "2-film" theory, first posited by Whitman (1923). In this representation, the mass and heat transfer (convective and conductive transfer, respectively) between the two phases is driven by the difference in concentration or temperature between the phases. At the interface, it is assumed that the concentrations are at a thermodynamic equilibrium, with the resistance to mass transfer being determined by the properties of the fluid and the thickness of the film. In many situations the mass transfer from one of the films dominates and it is possible to reduce the problem to a single film.

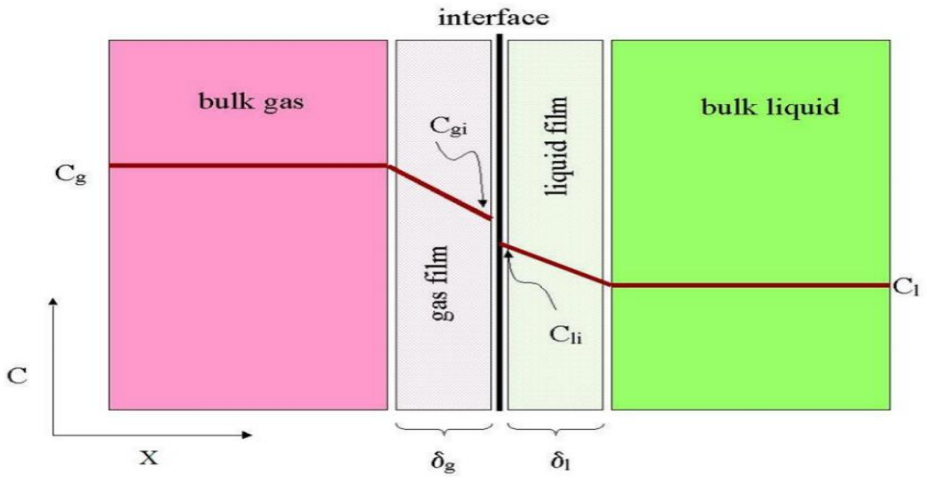

Figure 2.14: *Illustration of the 2-film theory (Source: Marmara University, 2016)*

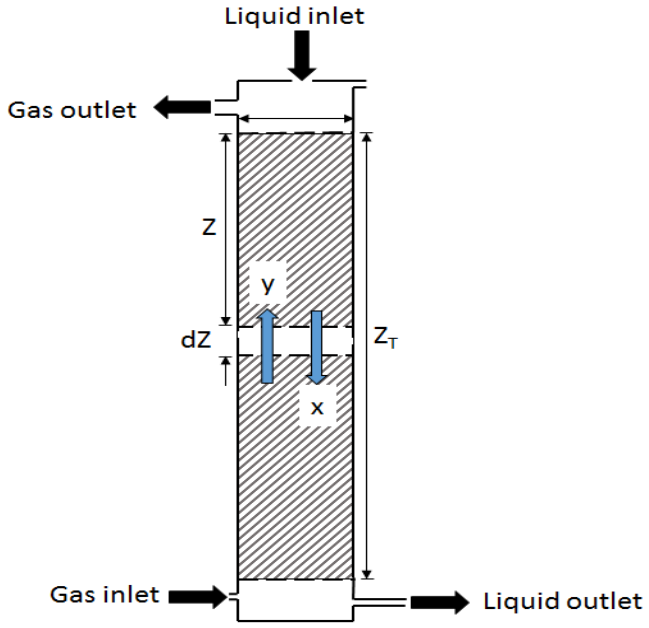

Figure 2.15: *Diagram of a packed column (Source: McCabe, et al., 2005)*

2.5.2.2 Packed Column Design

In so far as the design of packed columns is concerned, the literature is fairly sparse. While much work has been done on staged columns, packed columns are often described using approximations. The most common approach to designing gas-liquid packed columns has been through dividing the column into a number of equilibrium stages, with the concentration profiles determined by assuming the equilibrium state of the gas and liquid streams leaving each equivalent stage. The method, known as HETP (Height Equivalent to a Theoretical Plate), has no theoretical basis (Seader & Henley, 1998) and since the packed column is a continuously contacting unit, it is best treated as such. In addition to this, it has been shown that it is not necessarily correct to assume that all the theoretical stages are of

the same height. In this study only methods that allow for the column to be considered as a continuous contactor are considered. Figure 2.15, on the preceding page, illustrates a typical packed column and the best way in which these columns should be conceptualised so as to avoid the use of shortcut models and simplifying assumptions as much as possible.

In order to model the representation in Figure 2.15 it is necessary to consider the mass balances over each differential element (Equation 2.15).

$$y_i . G|_{z+\Delta z} - y_i . G|_z = N_i^G \Delta z \qquad (2.15)$$

Where G is the molar flowrate of the vapour stream, z is the height point along the column, y_i is the molar composition of component i in the vapour phase, and N_i^G is the molar flowrate/flux of the component into the liquid stream. Dividing Equation 2.15 by Δz and taking the limit of Δz to zero the equation becomes the differential equation (Equation 2.16):

$$\frac{d(y_i . G)}{dz} = N_i^G \qquad (2.16)$$

The same can be done for the liquid mass balance in order to obtain Equation 2.17:

$$\frac{d(x_i . L)}{dz} = N_i^L \qquad (2.17)$$

In Equation 2.17, L is the flowrate of the liquid stream, x_i is the molar composition of component i in the liquid phase, and N_i^L is the molar flowrate/flux of the component into the vapour stream. The full list of equations that would be required to be solved in order to represent the packed column is listed in Chapter 6, in detail, but for now it should just be noted that a method for solving differential equations is required in order to design packed columns in a correct and rigorous fashion.

Due to the nature of random packings it is not possible to avoid the use of empirical correlations entirely. Figure 2.16, on the following page, shows a picture of a typical random or dumped packing, known as Raschig rings. These rings are typically packed into the column and serve the purpose of increasing the interfacial area for the mass transfer to take place on. Many different commercial packings are available, including Raschig rings, Hiflow rings, Berl saddles, Pall rings, etc. The selection of the optimal packing size and type are of importance, as these determine the interfacial area within any of the differential height elements, as well the cost of the internals of the column, and the propensity to flooding within the column.

Flooding can occur within a column when the velocity of the gas exceeds a certain velocity that forces the random packing materials to rise within the column, becoming entrained in the gas stream and forced upwards and out of the column. The details of flooding considerations are included in Chapter 6.

Figure 2.16: *Raschig rings (Source: http://www.christycatalytics.com/carbon-cylindrical-rings)*

In order to avoid the use of the simplifying assumptions used in the HETP method, rigorous methods for the simulation and design of individual packed columns are considered. In rigorously modelling continuously contacting columns, a differential system of equations is necessary to be solved.

2.5.2.3 Solving Differential Equations

The systems of ordinary differential equations (ODEs) that are required to be solved in the analysis of packed columns can be classed as boundary value problems (BVPs) because their solutions and derivatives can be specified at at least one point. Boundary value problems usually take the form:

$$\frac{d^2y}{dx^2} = f\left(x, y, \frac{dy}{dx}\right) \qquad \text{where } x \in (a, b) \qquad (2.17)$$

These problems cannot be integrated directly and are therefore usually solved using approximation methods such as shooting methods. In shooting methods the BVP is converted into an initial value problem (IVP) and attempts are made to find the unknown boundary derivative through multiple iterations, normally through the use of a Newton's Method solver. This solution through shooting methods is very dependent upon the stability of the IVP, as well as the initial conditions.

Another commonly employed method is to use the finite difference method in order to solve BVPs. These methods are the most commonly used approaches in chemical engineering applications. This method uses a Taylor Series expansion to approximate the value of the derivative at a number of small increments along a discretised grid:

$$f'(a) \approx \frac{f(a+h) - f(a)}{h} \qquad (2.18)$$

Where the distance between each gridpoint, h, is chosen to be a small number. The smaller this number, the more accurate the approximations are likely to be. The treatment of the problem in this way results in a set of algebraic equations that can be easily solved, however the large number of gridpoints required to obtain accurate solutions means it is computationally expensive.

Typically these two methods are utilised to simulate packed columns, with the columns' internals and diameter set. The design of the column is thus done by a designer in iterative steps until the designer is satisfied that the column meets the demands of the process. In order to fully simultaneously optimise the system using rigorous NLP optimisation techniques, another method is required.

2.5.2.4 Orthogonal Collocation on Finite Elements

The sequential and shooting methods described above often require the use of an embedded differential and algebraic equation (DAE) solver. In engineering applications, normally these problems have a variety of unknowns, including differential state variables, algebraic variables, as well as independent variables that do not relate to differential elements and are unique to the specific system studied. In order to fully optimise such a system, it is possible to create a fully open formulation, where all of the equations are explicit, through discretising the problem into a number of finite elements and using a piecewise polynomial representation of the functions within each element.

Consider splitting the region into a number of elements i, known as finite elements, of length h_i, and applying an interpolating polynomial on a number of collocation points within each of the elements, as represented graphically in Figure 2.17. In doing this it is then possible to solve the DE, g, at these selected points along the column height, z, by using an interpolating polynomial such as the Lagrange interpolation polynomial.

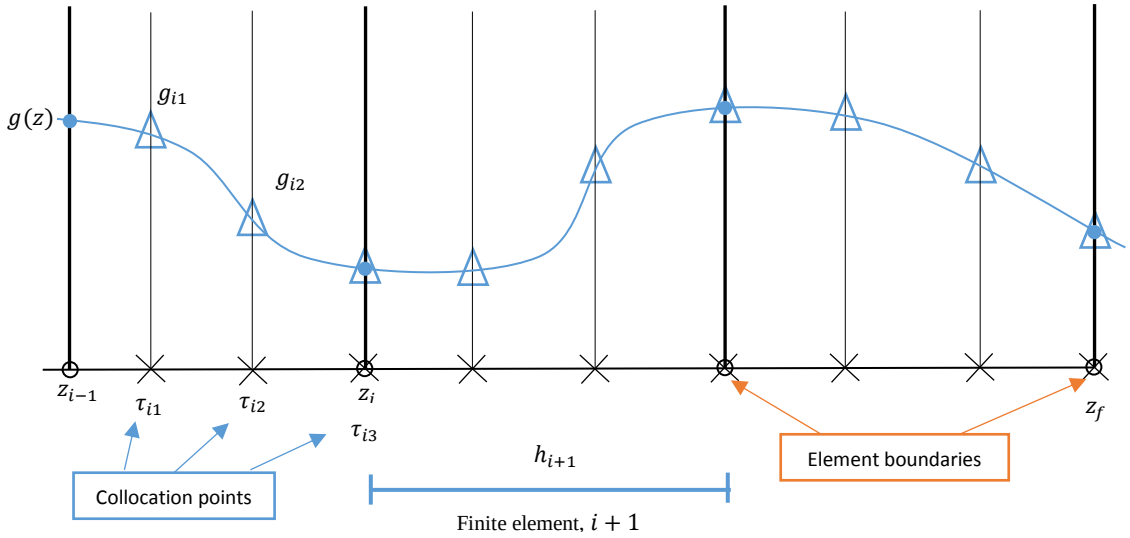

Figure 2.17: *graphical representation of collocation on finite elements. The triangles show dg/dz at collocation points and the circles show z at element boundaries, showing how continuity is retained.*

In Equation 2.19, below, the number of interpolation points is $K+1$ in element i.

$$z \in [z_{i-1}, z_i], \qquad \tau \in [0,1] \qquad \begin{cases} z = z_{i-1} + h_i \tau \\ g^K(z) = \sum_{j=0}^{K} \ell_j(\tau) g_{ij} \end{cases} \tag{2.19}$$

$$\ell_j(\tau) = \prod_{k=0,\neq j}^{K} \frac{(\tau - \tau_k)}{(\tau_j - \tau_k)}$$

Where $\tau_0 = 0$, $\tau_j < \tau_{j+1}$, $j = 0, \ldots, K-1$, and h_i is the length of an element. This interpolating polynomial is extremely valuable as it has the useful property that $g^K(z_{ij}) = g_{ij}$, at the collocation points, i.e. where $z_{ij} = z_{i-1} + h_i \tau_j$.

If one uses the Equation 2.19 to take first and second derivatives at the interpolation points one gets:

$$\frac{dg^K(z_i)}{dz} = \sum_{j=1}^{K} \mathrm{g}_{ij} \frac{d\ell_j(z_i)}{dz} \tag{2.20}$$

$$\frac{d^2 g^K(z_i)}{dz^2} = \sum_{j=1}^{K} \mathrm{g}_{ij} \frac{d^2 \ell_j(z_i)}{dz^2} \tag{2.21}$$

Vectors for the function, first, and second derivatives can then be defined as follows:

$$\boldsymbol{g_K} = [g_1, g_2, \dots, g_K]^T \qquad (2.22)$$

$$\boldsymbol{g'_k} = \left[\frac{dg^K(z_1)}{dz}, \frac{dg^K(z_2)}{dz}, \dots, \frac{dg^K(z_K)}{dz}\right]^T \qquad (2.23)$$

$$\boldsymbol{g''_k} = \left[\frac{d^2g^K(z_1)}{dz^2}, \frac{d^2g^K(z_2)}{dz^2}, \dots, \frac{d^2g^K(z_K)}{dz^2}\right]^T \qquad (2.24)$$

Rearrangement of these equations allows for the first and second derivative vectors to be defined as:

$$\boldsymbol{g'_k} = \boldsymbol{A}.\boldsymbol{g_K} \qquad (2.25)$$

$$\boldsymbol{g''_k} = \boldsymbol{B}.\boldsymbol{g_K} \qquad (2.26)$$

Where the coefficients can be determined through:

$$\boldsymbol{A} = \left(A_{ij} = \frac{d\ell_j(z_i)}{dz}; i, j = 1,2, \dots, K\right) \qquad (2.27)$$

$$\boldsymbol{B} = \left(B_{ij} = \frac{d^2\ell_j(z_i)}{dz^2}; i, j = 1,2, \dots, K\right) \qquad (2.27)$$

Typically, for NLP optimisation applications, either the shifted Gauss-Legendre or Radau roots are used to place the collocation points, as it has been shown that the roots of these orthogonal polynomials can provide desirable stability properties (Biegler, 2010). Radau points are preferred in NLP applications as they can allow for constraints to be set at the boundaries of each element, thus allowing for increased stability for high index DAEs (Biegler, 2007). Figure 2.17 demonstrates the application of Radau roots. Table 2.1, below, shows the collocation points for Legendre and Radau roots, and it can be noted that for Radau roots the final collocation point/root is always on the boundary of the next finite element. The placement of the collocation is of great importance to the accuracy of the solution, and the number of collocation points is of less importance (Rice & Do, 1995).

Table 2.1: *Shifted Gauss-Legendre and Radau Roots as collocation points*

Degree K	Legendre Roots	Radau Roots
1	0.50000	1.00000
2	0.211325 0.788675	0.333333 1.000000
3	0.112702 0.500000 0.887298	0.155051 0.644949 1.000000
4	0.069432 0.330009 0.669991 0.930568	0.088588 0.409467 0.787659 1.000000
5	0.046910 0.230765 0.500000 0.769235 0.953090	0.057104 0.276843 0.583590 0.860240 1.000000

Following on from Equation 2.19 above, the value of $g(z)$ can also be represented on a monomial basis for the differential profiles by Equation 2.28 with a Runge-Kutta-based representation, which is useful when forming the set of equations for an NLP:

$$g(z) = g_{i-1} + h_i \sum_{j=1}^{K} \Omega_j \left(\frac{z_i - z_{i-1}}{h_i}\right) \frac{dg(z)}{dz_{i,j}} \qquad (2.28)$$

Where g_{i-1} is the differential variable's value at the beginning of element i, h_i is the length of element i, z_i is the finite element position ($0<z_1<\ldots<z_{ne}=\Theta$), and $\frac{dg(z)}{dz_{i,j}}$ is the value of the first derivative in element i at the collocation point j, also referred to as τ. Ω_j is the interpolating polynomial of order K that satisfies:

$$\Omega_j(0) = 0 \text{ for } j = 1, 2, \ldots, K$$

$$\Omega'_j(\rho_r) = \delta_{j,r} \text{ for } j, r = 1, 2, \ldots, K$$

And ρ_r is the location of the rth collocation point in each element.

Continuity for the differential profiles at the boundaries of the elements is enforced by:

$$g_i = g_{i-1} + h_i \sum_{j=1}^{K} \Omega_j(1) \frac{dg}{dz_{i,j}} \qquad (2.29)$$

which ensures that differential variables are forced to be continuous throughout.

The algebraic profiles can be represented using a similar representation to the differential variables:

$$y(z) = \sum_{j=1}^{K} \Omega_j \left(\frac{z_i - z_{i-1}}{h_i}\right) \mathrm{y}_{i,j} \qquad (2.30)$$

Where $\mathrm{y}_{i,j}$ is the value of the algebraic variable in element i, at collocation point j. Ω_j is the Lagrangian polynomial of degree K where

$$\ell_j(\rho_r) = \delta_{j,r} \text{ for } j, r = 1, 2, \ldots., K$$

As has been shown above, it is possible to discretise the region of interest in a differential equation and solve the set of equations simultaneously by turning the DAE system into a set of algebraic equations. By doing this, NLP optimisation techniques can be directly applied to accurately approximate the differential equations, as well as optimise key variables within the process without the use of approximations that forego the solving of DEs. The method has been shown by a variety of authors to be sufficiently accurate, as well as having the advantage of being able to be solved in much faster times than other methods used in solving DAEs.

2.5.2.5 Application to Packed Columns

Many different approaches have been utilised in order to simulate the packed bed absorber system that is the focus of study in Chapter 6 of this thesis, however most of these studies have attempted to simulate a lab-scale unit accurately, without concern for unit optimisation. Brown and von Rosenberg (1963) was one of the first works to model the packed system as a continuum, when they simulated a multicomponent system at steady state. The work was done using analogue computers. The same system was then solved as a continuous system using a central difference method to solve the first order differential equations with split boundary conditions. Von Rosenberg and Hadi (1980) extended this approach by including non-linear equilibrium relations.

Arwikar and Sandall (1980) conducted experiments on binary systems with a Raschig ring packed column. They were able to eliminate end-effects by using differential height techniques. In the systems studied, the liquid phase was saturated, indicating that the liquid phase mass transfer offered no resistance in distillation systems, however the absorption

experiments indicated that there was liquid phase resistance that was small but not negligible.

Krishnamurphy and Taylor (1985) used mass and energy balances over a number of elements in terms of each phase in their simulation of packed absorbers, which they solved using Newton's method. Their model, which serves as the basis for Aspen's packed column models, makes use of non-equilibrium stage models that solve the mass and energy transfer rate equations simultaneously for each phase, as well as interface equilibrium equations. These equations are known as MESH (mass, equilibrium, entropy, enthalpy) equations. This method is still the predominant method used by most process engineers and requires that one of the boundary conditions is specified, as well as the diameter and height of the column, with the model solving for the concentration profiles and final concentrations.

A number of other studies also used a similar approach to simulate experimental set-ups or to gain insights into the physical process within the packed column. De Leye and Froment (1986) used rigorous modelling to simulate packed columns. Hüpen and Kenig (2004) simulated NO_x absorption in both packed and staged columns using a rigorous general rate-based model for reactive absorption that made use of Aspen Custom Modeler ™ and validated their work through the use of experiments. The rigorous model used experimentally-determined correlations from Billet and Schultes (1999) for the mass transfer coefficients, interfacial areas, and volumetric hold-up. Kloker, et al. (2005) designed a rigorous rate-based approach for simulating reactive separation systems and implemented it into a process simulator. The models were rigorously verified in a pilot plant.

Due to the continuous nature of the packed columns, as described above, it is surprising that fairly little work exists on the application of OCFE to packed columns. Steiner, et al. (1978) divided the column into 10 elements and used a finite difference method that made use of a general overall mass transfer coefficient, however the first use of orthogonal collocation for packed column simulation was by Srivastava and Joseph (1984). They studied the same system as von Rosenberg and Hadi (1980) but used polynomial approximation techniques and made use of the 2-film mass transfer theory. Their solution was found to be accurate, even with fewer than 4 collocation points and without the use of finite elements. Their study was the first to show that the collocation method greatly decreased the number of equations required to be solved in comparison with the finite difference method. Their model had fixed packing parameters, column diameters (and therefore velocities), and solved for the packing height required.

The detailed packed absorber designs from Kenig, Gorak, and colleagues at the University of Dortmund were utilized by Algusane, et al. (2005) in their framework for the synthesis of reactive absorption columns. There model used orthogonal collocation (OC) in a novel framework for the synthesis of optimal staged reactive absorption columns. By utilizing OC with a novel Generalised Modular Framework (GMF) they managed to greatly decrease the model order and still retain the stage-wise nature of the staged column through application of the polynomial approximations being applied at specific points along the column. The model was shown to very closely resemble the detailed, stage-wise model, while only using 4 collocation points within each module.

Few attempts at optimisation of the design of packed columns have been made, with the vast majority of these studies focusing either on accurately simulating columns to match experimental data, or with the focus on dynamic behaviour and using model predictive control to optimally control a column.

Karacan, et al. (1998) was the first to simulate both the dynamic and steady-state behaviour in a packed column using experiments to validate the approach. Their approach used OCFE on a two-film back-mixing model and used both Legendre and Jacobi polynomials to test their effectiveness. The results show that OCFE is effective in predicting the highly nonlinear column behaviour and showed that, for his system, 7 Legendre polynomial collocation points gave good approximations for the column, however only 4 points provided accurate solutions for orthogonal collocation on finite elements with only 2 finite elements.

Torres, et al. (2000) compared the results between a rigorous non-equilibrium model and a reduced order collocation-based model for packed column separation based on that presented by Srivastava and Joseph (1984). Their model found that the reduced order model sufficiently modelled the system with regards to the concentrations from the products, however sometimes performed poorly in predicting the concentration profiles along the columns.

As far as the optimisation of column design is concerned, the current author is only aware of the models of Mores, et al. (2012). They used different approaches to optimize a packed column for CO_2 absorption using a monoethanolamine solution. The study found that a simplified rate-based model was sufficient in modelling the system accurately and then used this model in GAMS to optimize the individual column. Non-ideal behaviour was found to have little effect on the solutions obtained. The method did not model the column as a continuum, rather splitting the column into a number of sections and then calculating non-

equilibrium efficiencies in each stage. They tested various effective mass transfer area approximations and found Onda's (1968) to be the most suitable. The model found interesting absorber configurations with variable diameter, or series exchangers with variable diameters, but kept the packing characteristics fixed and used an objective function related to maximizing the CO_2 absorbed per total packing volume, rather than one based on cost.

From this literature survey it can be observed that there exists a lack of research with regards to using rigorous models and NLP optimisation techniques to find the optimal packed column design. The details of the approach developed to solve for optimal packed columns in this thesis is found in Chapter 6.

2.6. Toward Complete Simultaneous Network Design

Throughout the literature review thus far, it has been shown that there are two separate tasks that are intrinsically linked, but have thus far lacked any clear connection in the process synthesis field. These two tasks are the detailed unit optimisations on the one hand that can make use of either simulations, or optimisation strategies, and that often involve a large number of variables that include detailed thermodynamic information. On the other hand we have network optimisation which generally involves simplifying the problem into formulations that are as convex as possible with a large number of assumptions and fixed parameters. The perfect optimisation procedure, as has been demonstrated would involve both of these representations in combination so that changes that result from detailed unit design can be included at the level of the network optimisation, because the solution to each problem relies on the solution of the other. This has been mostly avoided in HENS and MENS with authors mostly concentrating on either simplifying assumptions in order to optimise the network, or attempting to include detailed simulations in the designs of the individual units. Some authors have attempted to combine these two separate optimisation problems to a degree of success in other areas of process synthesis.

2.6.1 Combined Optimisation and Simulation in Process Flowsheets

In an effort to use rigorous simulation models within an optimisation framework, Diwekar, Grossmann, and Rubin (1990) proposed the use of the commercially available and commonly used process simulator ASPEN in a decomposition strategy similar to that of Kocis and Grossmann (1989), described in 2.4.1.4. In their optimisation strategy the MILP solver and NLP solver are included as separate unit operation blocks in ASPEN. The method first postulates the superstructure and then decomposes this into an initial flowsheet with a subsystem of non-existing units which will be suboptimised using a Lagrangean scheme that provides linear approximations for the whole superstructure. NLP optimisation of a selected flowsheet and its subsystems takes place in the inner loop, with the Lagrangean multipliers from this initial flowsheet passed onto the subsystem to perform the optimisation. The resulting gradients, objective function, and Lagrangean multipliers from the selected flowsheet and subsystems are then sent to the master block in ASPEN. This master block changes the master problem internally through the ER strategy, with the information obtained in the inner loop. A new flowsheet topology results from this problem and a new NLP optimisation is undertaken, with the iteration stopping only when no improvement is seen in the objective function (Diwekar, et al., 1990).

Meshing a sequential modular simulator like ASPEN with an MINLP equation-oriented optimisation environment, like GAMS, cannot provide satisfactory results. This is because the design equations and constraints that are usually explicitly stated in an environment like GAMS, are actually only included implicitly as the modular simulator only provides the input and output information. The detailed design equations are therefore not available to the optimiser. A strategy is therefore required to provide constraint linearisations. The strategy proposed by Diwekar, et al. (1990) involves separating the continuous variable vector into 2 categories, input and output variables and uses these to construct the linearisations.

Diaz and Bandoni (1996) applied a similar synthesis strategy to make use of rigorous simulation along with MINLP optimisation strategies for a large-scale chemical plant. They applied the OA technique and found that significant savings could be made when compared with NLP techniques, even though there is no way of determining whether the solution found is globally optimal. They also noted the importance of starting at many different initial points as the complex non-linear equations within the simulators may lead to non-

convexities that can result in suboptimal solutions and by providing multiple starting points more potential local optima can be evaluated.

Their approach to obtain the linearisations for the implicit variable calculations differed from Diwekar, et al. (1990) in that gradient information was obtained numerically by finite differences using the forward difference formula, using perturbations of $0.001x_i$. The use of this estimation requires large CPU times because the NLP requires a number of entire process simulations for each individual variable (Diaz & Bandoni, 1996). Furthermore, these linearisations will only be valid over narrow bands within the operating regime that is first selected.

Brunet, et al. (2012), used a multi-objective approach to the design of absorption systems using a similar approach to Diaz and Bandoni (1996). In this formulation ASPEN was again used as the simulator with implicit equations and a Pareto curve was generated including an environmental objective function. A key difference in their formulation was that, at each iteration, the simulation must converge for each set of design variables and this was handled by the inclusion of slack variables and an exact penalty to the objective function in order to handle infeasible solutions.

With these approaches it can be seen that it is extremely challenging to include detailed models in combination with a large MINLP flowsheet optimisation. The most challenging aspect involves the use of binary variables to decide between different process configurations and it has thus been far more common to model and optimise fixed topologies for plants. When detailed information and rigorous bounding and initialisation strategies are employed it is possible to solve NLP formulations that contain several hundreds of thousands of variables. Biegler and his associates at Carnegie Mellon University have solved many systems using NLP formulations with the inclusion of detailed thermodynamic considerations, however the inclusion of binary variables still makes the solution of networks, especially large ones that cannot be simplified to NLP formulations, a massive challenge for the process synthesis community (Biegler, 2010). Additionally, these large NLP models cannot be guaranteed to converge to globally optimal solutions, and therefore often need to be solved using a wide variety of initial conditions.

2.6.2 HENS and MENS with Detailed Unit Design

While attempts have been made to merge process flowsheet optimisation with detailed models of individual units and processes, there has been very little research on attempting to include detailed unit design into HENS and MENS. As the previous sections have outlined, the majority of optimisation attempts have either concentrated solely on either network generation or individual unit design. There have however been a few attempts at including more detailed aspects of the design into the network optimisation. The need for including detailed designs at the network generation is vital as many of the assumptions made at the network generation stage might not hold at the individual unit design as the equations that have been used at the network level are simplified in order for the large number of equations and discrete and continuous variables to be solved to close to optimality. Much of the literature reviewed in this section is also reviewed in detail in Chapters 4, 5, and 6.

A modified SWS optimisation by Soršak and Kravanja (2002) allowed for the simultaneous consideration of different types of exchanger. The modifications implemented are in the form of disjunctions included at each possible heat exchanger match in the superstructure, with additional constraints included which ensure that feasible temperature distributions are allowed for every type of exchanger. These added nonlinear constraints required the development of a new MINLP solution strategy whereby an initialisation scheme is postulated which is shown to halve the time taken to solve the MILP master problem in the modified OA/ER algorithm. The individual exchanger calculations are still of the form used by other authors, and parameters such as the overall heat transfer coefficients and the consideration of multiple shells and F_T correction factors are excluded.

Frausto-Hernandez, et al. (2003) used Jegede and Polley's (1992) pressure relationship that relates the individual heat transfer coefficients and exchanger area to formulate an MINLP SYNHEAT model that considers pressure drop simultaneously with area. The method uses allowable pressure drops for each stream and calculates different overall stream heat transfer coefficients based simultaneously on exchanger area and total pressure drop of the stream. The method was shown to be highly non-convex and therefore globally optimal solutions were not guaranteed. In addition, only single pass shells and tubes were considered.

Mizutani, et al. (2003b) couples the Yee and Grossmann (1990) SWS superstructure with the individual exchanger model discussed in Section 2.5.1.2 and in Chapter 4 in a novel formulation that involves disjunctions in the topology selection as well as the detailed heat exchanger design. The model contains 3 levels of discrete choices: first are network topology decisions based on the SYNHEAT model, second are the design decisions for the individual units and third are the disjunctions that relate equations to flow regimes based on Reynold's number. The method used the upper bounds for heat transfer coefficients and LMTD in the MILP master problem in order to underestimate the objective function for unselected matches. Major problems with the method are that only single-pass non-TEMA exchangers are considered, pumping costs are not considered in the topology selection, and that multiple shells are not considered. The fact that only single pass and single shell exchangers are considered can potentially be very limiting as it is standard design practice to consider multiple tube and shell passes and that the pumping costs will be unrealistically low for cases of single shell and single tube passes.

In a similar procedure to the method of Mizutani, et al. (2003b), Ravagnani and Caballero (2007b) used their TEMA model with considerations of number of shells and F_T correction factors in conjunction with the SWS model in a bi-level decomposition. Their methodology considers stream splitting and constant heat transfer coefficients in the first SYNHEAT MINLP. The initial HEN is then designed in detail using Ravagnani and Cabellero's (2007a) MINLP formulation of individual HE optimisation. Using these detailed exchangers, a global TAC considering pumping and the new heat transfer coefficients is obtained. The heat transfer coefficients obtained from this detailed design are then inputted into the MINLP SYNHEAT model and it is re-run. If the network is the same as the previous solution or the design gives a worse global TAC the procedure is stopped and the optimal network is obtained. The authors admit that this procedure is heuristically based and is therefore not necessarily the global optimum. Furthermore, it is the present author's observation that the addition of new heat transfer coefficients alone does not necessarily allow for a fully optimal network structure to be generated. This is because important considerations that are dependent on the topology are not applied to the MINLP SYNHEAT topology optimisation. Aspects of the design that can have an effect on the topology that are excluded from the topology optimisation include the effect of pressure drops, the F_T correction factor, and the influence of multiple shells. A further observation on the technique used by Ravagnani and Caballero (2007b) is that the individually optimised exchangers never have a triangular tube arrangement chosen in the optimal solution. This is most likely due to the shell-side pressure

drops that are smaller with square arrangements. In industry triangular tube arrangements are common as they give very good heat transfer, and therefore the objective of the individual heat exchanger optimisations may be poorly constructed.

Wang, et al. (2012) made use of a unique algorithm for the retrofit of HENs based on heat transfer enhancement. Their algorithm used 4 heuristic rules to determine HEN bottlenecks and identify which heat exchangers would benefit from heat transfer enhancements. The method included a new way of modelling the shell-side pressure drop and heat transfer co-efficient that is simplified and provides results that compare closely with commonly applied commercial software, such as HTRI ®.

To the best of the knowledge of this author there seems to have been no real attempts at including greater design details into the standard MENS formulations. Isafiade and Short (2016), as discussed in Section 2.4.3.1, included extra details of the design into the MINLP formulation by allowing for the consideration of the diameter as a process variable. This resulted in the necessity to include other aspects such as the velocities and their effects on the mass transfer coefficients. The model was used in both single and multi-period mass transfer, however due to the inclusion of these highly nonlinear extra terms, the solutions obtained, even for very small examples, required special initialisation and bounding strategies and the resulting networks could not be guaranteed to be globally optimal. Additionally, the model had no way of including flooding considerations at the level of the MINLP, and so an additional step was required in order to check the flooding considerations. Once the flooding conditions were checked, another iteration of the MINLP was required in order to be run with the updated packing.

Unfortunately none of these methods really provide the MINLP topology optimisation with greatly improved access to the information required to actually help guide the design. Including more detailed models into the MINLP optimisation should be avoided as the highly non-convex systems that are already being utilised have been shown to be difficult to solve, and as problems get larger it becomes increasingly difficult to find feasible and even locally optimal solutions, even with special initialisation procedures. By only updating one parameter in every iteration, as was done for the packing sizes by Isafiade and Short (2016) for MENS and for the overall heat transfer coefficients by Mizutani, et al. (2003b) and Ravagnani and Caballero (2007b), it greatly simplifies the impacts of the other detailed aspects of the designs that will influence the solution if the network generated were to be reproduced in a functioning industrial environment. The methodology presented in Chapter

3 and applied to various problems in the subsequent chapters presents a more systematised and rigorous way of including detailed design aspects into the MINLP network optimisation without the requirement for increasing the non-convexity of the problem, while also ensuring that the resulting network is still accurate in accordance with detailed design methods.

Chapter 3

Methodology

Chapter 3: Methodology

3.1 Introduction

This chapter presents the methodology used for the synthesis of heat and mass exchange networks developed in this thesis. As has been evident throughout the literature review in Chapter 2, a large gap exists between the models used in network synthesis and those used in the detailed design of the individual units that comprises the network. The approximations used to size the individual units, as well as the static stream parameters that are used to describe the heat and mass transfer coefficients and the log mean difference approximations that are required in order to size the units often result in networks that are optimal in terms of the sets of equations and variables, but fail to be optimal once the units are designed using more appropriate representations that involve detailed unit design and simulation.

A key problem in trying to find formulations for the network optimisation that better represent the actual design equations needed for accurate designs is the fact that MINLP solvers are limited. Even with the shortcut models used in modern synthesis techniques, local optima are common and it is often not easy to find feasible solutions in problems with large numbers of streams. It is therefore not advantageous to increase the non-linearity or size of the current MINLP formulations, as the attempts at including more detailed design aspects have resulted in extremely difficult problems to solve that require taxing manual initialisation and bounding strategies that can often still lead to only locally optimal solutions. This thesis presents a way to synthesise HENs and MENs without the addition of new non-convex equations or binary variables into standard superstructure-based MINLP models by decomposing the design procedure into a network topology optimisation stage and detailed unit design stage that are connected through a novel algorithm that allows for the MINLP to be updated with a series of corrections in order for the MINLP to be guided by these more rigorous individual unit models.

3.2 Decomposition Strategy

Since the MINLP formulations that have been used up until now are difficult to solve to global optimality and often require special initialisation and bounding strategies in order to find feasible solutions, it is better not to attempt to increase the non-convexity of current formulations. In order to include the detailed design equations explicitly into the MINLP formulation new non-linear equations, continuous and binary variables, and bilinear equations would be required, and these are to be avoided. For this reason the approach of this thesis is to decompose the problem into two subproblems, an MINLP topology optimisation and a unit design step.

The initial MINLP will be formulated in the same way as the other superstructure-based methods for HENS and MENS, however it will be supplemented with a number of sets of new parameters. These parameters, referred to throughout the text as correction factors, are placed in the objective function, affecting the selection of specific topologies by correcting the solutions of the MINLP towards those obtained by the detailed models. Since the correction factors are derived externally to the model, they do not increase the size or non-convexity of the MINLP formulation. This allows for the possibility to include information from the detailed unit designs implicitly, even allowing for the inclusion of extra design information that has yet to be included in any meaningful way into the network synthesis tasks previously. Examples of new information that is included in the case studies includes, pressure drops and number of shells in HENS, and diameters and flooding considerations in MENS.

Since the MINLP network topology optimisation generates networks that have the correct mass and energy balances, temperature and composition differences, mass and heat transferrals, and the selection of the matches; these can all be inputted into the subsequent detailed design section. The detailed unit design section can make use of any method necessary in order to design the specific units that would be required to carry out the respective duties given by the MINLP. This study has made use of a detailed heuristic/empirical data design method for HENS, which is detailed in Chapters 4, and 5, and a novel detailed OCFE NLP optimisation for the determination of optimal packed columns for the MENS, detailed in Chapter 6. It is possible, however, according to the method

presented here, to use any design technique at the designers' disposal for this step, including commercial simulation software packages such as ASPEN, HYSIS, HTRI, etc.

The resulting units from the detailed designs, that have used more accurate and detailed equations for the unit sizes, can then be compared to the solutions from shortcut models used in the MINLP. The differences between these solutions can be used to determine the correction factors, which are then inputted into the MINLP for the subsequent iteration of the algorithm. Figure 3.1, below, displays the methodology and decomposition strategy graphically, demonstrating how the two steps are linked.

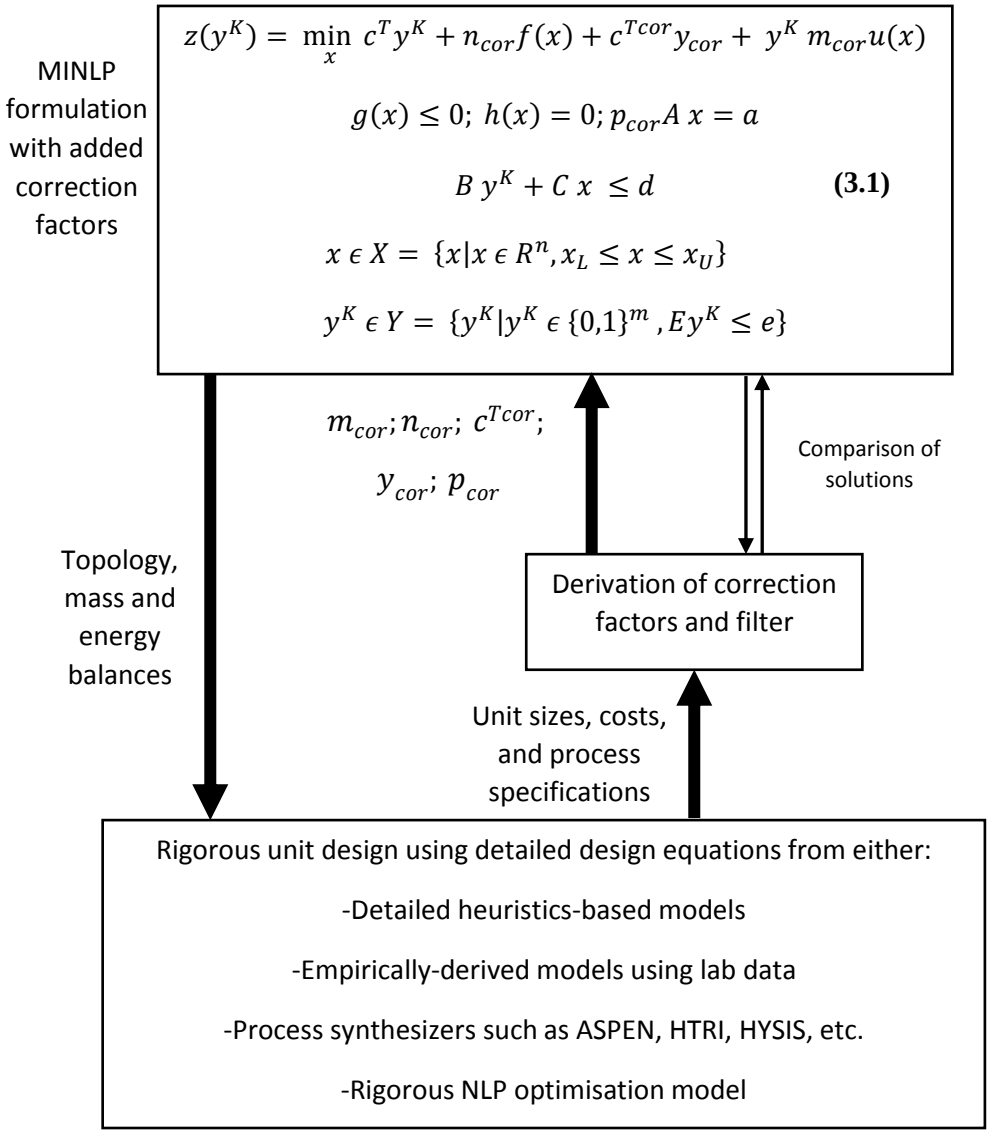

Figure 3.1: *Graphical representation of the decomposition strategy for this thesis*

In Figure 3.1 the standard MINLP formulation presented in Chapter 2 as Equation 2.14 is modified to include the problem-specific correction factors (Equation 3.1). The general correction factors here represent the following: n_{cor} are correction factors to the unit sizes (i.e. column heights and heat exchanger areas), y_{cor} are relaxed binary parameters that can signal the existence of extra aspects to some units, (e.g. does the heat exchanger require an extra exchanger in a certain period of operation?), c^{Tcor} is a cost associated with any extra additions to the network, m_{cor} are correction factors associated with continuous variables in functions that are dependent on the existence of a specific match (e.g. pressure drops and the associated cost function in heat exchangers), and finally p_{cor} which represents the changes to the problem-specific stream data as a result of a particular match (e.g. heat and mass transfer coefficients). It should be noted that all of these correction factors are specific for a particular match within a specific heat/mass exchange interval within the interval-based superstructure to be utilised. However, it should be known that this does not imply that the method cannot be applied to other superstructure based synthesis methods such as that of Huang and Karimi's (2013) exchanger-centric model or that MINLP hyperstructure approach of Papalexandri and Pistikopoulos (1994). In certain case studies some of these aforementioned correction factors may be excluded as they are dependent on the specific examples to be studied and the information which the designer wishes to consider. The details of what these correction factors represent for each case study are presented in detail in the methodology sections of Chapters 4, 5, and 6.

The specifics of how the algorithm proceeds, how the optimal solution is found, and the way in which the correction factors are derived are presented in Section 3.3.

3.3 Algorithm

The algorithm, represented graphically in Figure 3.2 on the next page, is as follows. Once the newly formulated MINLP is run and a network is obtained, the mass and energy balances, as well as the topology are then inputted into the detailed design stage so that the rigorous unit design can take place. Once these units are designed in detail to perform the required duties, the costs of the detailed network are obtained. It is vital that the costing functions of the objective function of the MINLP utilise the same costing functions as the unit designs, as the optimal solution should be based on detailed costing functions, as well as the details of the detailed unit designs. The non-convexity of the costing functions, noted in Chapter 2, is less of an issue than many of the other non-convex functions that may reside in the main formulation, as they lie within the objective function, where they are far simpler for the solvers to manage (Yee and Grossmann, 1990).

The objective functions from the MINLP and detailed network need to be compared at the next step. The detailed network design, aims to use detailed equations and deterministic solvers or heuristics to find optimal or near-optimal individual unit designs based on the mass and energy balances from the MINLP. This is with respect to the same costing equations used in the MINLP. There will be discrepancies between the two objective functions (MINLP and detailed unit design) at this stage, and the objective function that will form the basis of an optimal solution is the one resulting from the detailed network, because this objective function will be a better representation of the real costs of the network as fewer assumptions regarding the unit designs are made. The solution to this network which is designed in detail is saved as the best current solution if it is better than the previously saved objective function of the previously best detailed designs.

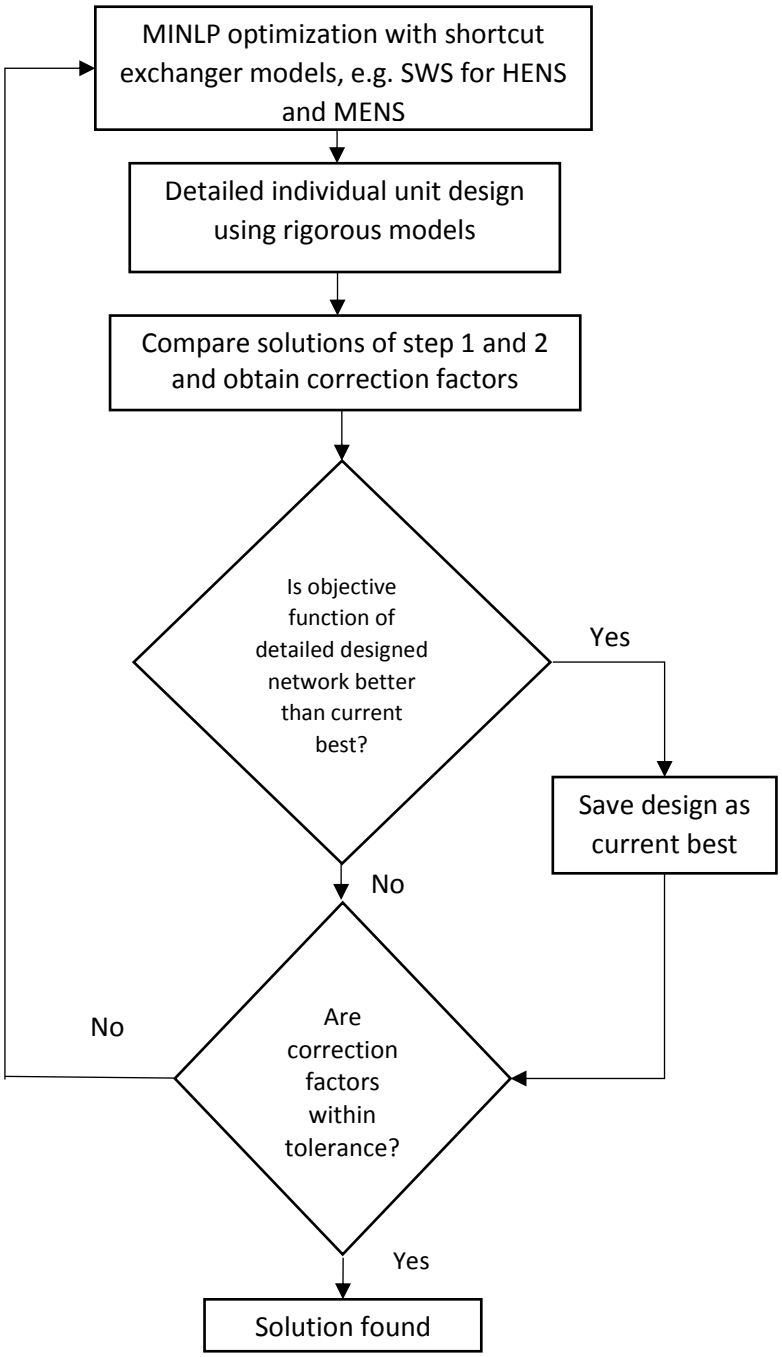

Figure 3.2: *Representation of the iterative procedure used in the algorithm*

The correction factors can now be determined. The correction factors are designed in such a way so that the value of the objective function of the MINLP will eventually converge to the same objective function value as the detailed design. This is done by multiplying the specific matches' areas/sizes, heat and mass transfer coefficients, costing specifications, etc., by the respective correction factors. For example, if the area obtained from the MINLP for the match of hot process stream 1 with cold process stream 2 in interval 3 (i.e. match '1','2','3') is 10 m^2, but after doing a detailed design using the temperatures and heat load

together with detailed heuristics, an area of 10.3 m^2 is obtained. This then means that the required correction factor for the area of that match ('1','2','3') is 1.03.

The correction factors are obtained for the process specifications, the existence of extra features that may exist, and the other design features that will affect the overall cost and proper functioning of the network that the MINLP formulation has not been able to anticipate. In order for the solution space to not be altered too drastically, a filter is applied to the amount of change that a correction factor can undertake between subsequent iterations. This filter value is chosen arbitrarily, however the smaller the value, the more potential network topologies can be unearthed during the iterative process, but at the cost of a large increase in the number of iterations required for convergence. This value is chosen as 5 % and 10 % during this thesis, with detailed values for all correction factors and their implementation shown in the respective chapters to follow. An example of this "filtering" effect can be illustrated as follows: if the mass transfer coefficient obtained from the MINLP for the match of rich process stream 2 with lean process stream 1 in interval 3 (i.e. match '2','1','3') is 0.05 kg/s/m^3, but the detailed design suggests it is 0.032 kg/s/m^3, then the required correction factor for the mass transfer coefficient ('2','1','3') is 0.95, not the unfiltered value of 0.64. The reasons for this filtering and its effect on the convergence is mentioned briefly in Section 3.4, with greater detail provided in the respective chapters with more concrete example problems.

Once these correction factors are obtained and implemented into the MINLP, the MINLP is re-run. The resulting network from the MINLP may have changed topology, or may have only changed split flow arrangements, or may have not changed at all, however the objective function value will now more closely mimic the results obtained from the detailed designs. The resulting network is re-designed in detail, with new correction factors obtained. At every iteration, the detailed network objective function is of the most interest as it represents the actual estimated cost for the network, as opposed to the MINLP objective which is a more loose approximation. This value is saved and re-written every time a better network is obtained with regards to the detailed costs.

The algorithm repeats until either the correction factors are unchanged between two consecutive runs, or until a maximum number of iterations, set by the user, is reached. When the correction factors have converged upon the same value (or do not change by less than some tolerance between runs) the objective value of the detailed network and the MINLP should be the same. It is found that the converged solution and the globally optimal solution may not be the same, and the reasons for this are explained in the chapters where the case

studies are run, but it can be shown that this is due to the presence of local optima in the MINLP optimisation. The optimal network solution is thus selected as the detailed design that gave the lowest TAC throughout all the iterations.

3.4 Initialisations and Convergence

Initialisations are of great importance in the proposed method. As all of the correction factors are parameters that are implicit, i.e. come from outside of the optimisation, it is important to select values for these parameters that will not exclude potentially optimal networks. This can be done by selecting parameter values that will underestimate the MINLP's objective function. By doing this, subsequent iterations of the procedure will include the underestimated topological choices, ensuring that a large number of networks are evaluated throughout the algorithm. These parameter choices are vital, and include both the correction factors themselves as well as the process parameters such as the diameters of the packed columns and mass transfer coefficients in Chapter 6 and the heat transfer coefficients and pressure drops in Chapter 4. A balance should be struck between the amount of underestimation of the parameters and the filtering of the correction factors, i.e. the limiting of the change of the correction factors between iterations. This balance is important because if the parameters are underestimated by too large a magnitude, then it will take a large number of iterations for the algorithm to reach the correct parameter values. Selecting a high filtering value, by contrast, will speed up the convergence towards these values, however at the cost of possibly excluding potential solutions that lie between the parameter values. Another factor that should be noted is that often the correction factors are multiplied together, or compounded, and if no filtering value is selected then this could greatly overestimate the effect of the corrections upon the final solution. This effect can be seen in the case studies shown in the subsequent chapters.

Since it is important to obtain good initialisations that are neither too far away from potential actual values, but also at the right level to underestimate the objective function, it is advised to undertake an exploratory run of the MINLP and detailed unit designs in order to obtain the initialisations. By using estimates for the parameters initially and then running the MINLP and proceeding with the design, it is possible to determine the likely region

wherein the parameters will lie and use this to choose values that will underestimate the objective function.

Convergence of the problem to a point where the correction factors do not change between consecutive runs and the objective function values of the detailed network and MINLP are the same cannot be guaranteed. This is due to the presence of the binary variables, which may just switch between different equivalent networks ad infinitum, as well as stream splitting, which may result in small changes of the split flowrates between iterations, causing the correction factors to oscillate between iterations. Additionally, since it is not possible to guarantee that a global optimum is achieved by the MINLP, even through the use of mostly non-convex formulations in this thesis, it is not possible to guarantee that the final converged solution, if one is obtained, will be the globally optimum solution. In fact, in the chapters to follow, it can be seen that this was rarely the case. The reason for this is one of the great strengths of the proposed methodology. The fact that the correction factors are constantly changing between iterations shifts the starting points/initialisations of the MINLP. These small perturbations can have large effects on the directions of search for the solvers and can thus be very helpful in not only guiding the MINLP toward more realistic designs, but also in unearthing new topological structures that would have been overlooked by the solvers previously. The new algorithm presented here can thus greatly increase the chances of finding a globally optimal solution through this structured multi-start procedure. While each iteration of the MINLP is at least locally optimal, the method presented here is shown to often find structures that would have otherwise been only found through lucky initialisations of the MINLP, rather than through the structured multi-start perturbations used here.

The strengths of the method are therefore that the chances of globally optimal solutions to the MINLP are greatly increased by the structured generation and analysis of a large number of structures and the fact that the inclusion of detailed designs that are used to guide and lead the MINLP topology optimisation towards more realistic and detailed designs without increasing the complexity of the MINLP formulation can lead to a guarantee that the generated networks can be implemented in real-life applications.

The chapters to follow include the specific implementation of the general methodology laid out in this chapter. Due to the nature and specificity of each application, the full published relevant papers are included, which all include their own literature reviews, relevant model formulations, additional problem-specific correction parameters included into the objective functions, and the results as applied to the various case studies which justify and confirm

the validity and usefulness of the approach. Chapter 4 presents the application to HENS, Chapter 5 extends the approach to multi-period HENS, and Chapter 6 shows the effectiveness of the method when applied to MENS, including a novel OCFE NLP packed column optimisation.

Chapter 4

Heat Exchanger Network Synthesis

Chapter 4: Heat Exchanger Network Synthesis

This chapter is a verbatim reproduction of a paper published in the 144th *Chemical Engineering Science* Journal on 4 February 2016 entitled "Synthesis of heat exchanger networks using mathematical programming and heuristics in a two-step optimisation procedure with detailed exchanger design" (Short, et al., 2016a). The only changes are cosmetic so as to retain the requirements for the thesis, which state that "there should be consistent format style throughout the thesis". The work is the sole work of the current author, with input and help from the co-authors not more than supervisory and whose valuable input and help in producing the paper are duly acknowledged. The chapter details the ways in which the methodology discussed in Chapter 3 is applied to the example of single period heat exchanger network design. The paper summarises the key literature developments, some of which is detailed in Chapter 2 of the thesis and elaborates on the contributions of the paper and how the new method is able to generate unique solutions that are of a higher quality than current optimisation techniques. The paper lists all of the relevant equations used for both the MINLP formulation and the detailed heat exchanger designs based on the heat and mass balances derived from the topology optimisation. It also shows the way that these detailed designs are utilised in the algorithm and how they force the MINLP optimisation to give more realistic solutions that can ultimately lead to better network designs. The methodology is applied to two case studies and compared to the solutions of other authors, showing the advantages of the approach over other techniques.

Synthesis of Heat Exchanger Networks Using Mathematical Programming and Heuristics in a Two-Step Optimisation Procedure with Detailed Exchanger Design

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Abstract: This study makes use of a novel methodology for the synthesis of heat exchanger networks, which is aimed at overcoming the shortcomings associated with the use of shortcut models to represent individual exchangers in the synthesis network. The new approach entails the use of a number of correction factors to get networks which are based on the use of shortcut models, such as the mixed integer non-linear programming (MINLP) stage-wise superstructure (SWS) of Yee and Grossmann (1990) to more closely represent physically achievable heat exchangers and ensure that the MINLP network topology optimisation step of these models converge on a real design, rather than an approximated one. In this paper, the SWS formulation is used for the generation of an initial network after which its objective function is modified to include the correction factors that force its objective function towards the cost of a network whose individual exchangers are designed using methods such as Bell-Delaware and heuristics. The modified objective function includes parameters that modify the areas obtained by the shortcut based MINLP model so as to more closely represent the areas obtained by the detailed models and also includes a novel method for including the number of shells required for each exchanger duty. The correction factors account for pressure drops, F_t correction factors, number of shells, TEMA considerations, and changes to the overall heat transfer co-efficient of each stream match. The methodology is applied to two examples and the solutions are comparable with other solutions obtained in literature and were shown to produce good solutions. The reason that the method is effective is because many potential networks are evaluated during the iterative procedure and the best network, based on the detailed exchanger designs, is chosen. In this way it is possible to use the detailed exchanger designs to "guide" the MINLP optimisation towards more realistic networks and also to generate many different potential networks.

Nomenclature

AC	Cost associated with area ($/m^2)
AE	Annualisation factor for area costs
C_p	Fluid heat capacity (J/kg)
CF	Fixed cost associated with purchase of a shell
CUC	Cost of cold utility ($/kW)
$DelPc_{j,k}$	Pressure drop of cold stream j across interval k
$DelPh_{i,k}$	Pressure drop of hot stream i across interval k
F_T	Correction factor by taking multiple tube passes into account
HEN	Heat exchanger network
HUC	Cost of hot utility ($/kW)
k	Conductive heat transfer coefficient of the fluid (W/mK)
LMTD	log mean temperature difference
MINLP	Mixed-integer nonlinear program
NLP	Nonlinear program
$NSP_{i,j,k}$	Number of shell passes for the exchanger i, j, k
Nu_i	Nusselt number
PC	Cost associated with pumping ($/m^{3}Pa)
Pr	Prandtl number
qc_i	Heat transferred from cold utility to hot process stream *i* (kW)
qh_j	Heat transferred from hot utility to cold process stream *j* (kW)
r_d	Fouling factor associated with fluid (W/mK)
SWS	Stage-wise superstructure
T	Temperature
TEMA	Tubular Exchangers Manufacturers Association
U_0	Overall heat transfer coefficient
VolFc	Volumetric flow-rate of cold stream

VolFh	volumetric flowrate of hot stream
$z_{i,j,k}$	Binary variable associated with the existence of an exchanger between the hot process stream i and cold process stream j in interval k
zcu_i	Binary variable associated with the existence of an exchanger between hot process stream i and the cold utility
zhu_j	Binary variable associated with the existence of an exchanger between cold process stream j and the hot utility
μ	Fluid viscosity (kg/m.s)
ρ	Fluid density (kg/m^3)

4.1 Introduction

Industry is increasingly looking at ways to reduce energy usage. Instability and rising prices of fossil-based fuels, due to geopolitical instability and rising extraction costs, as well as pressure from environmental groups and tax incentives from governments to reduce environmental footprints has driven research into producing more energy-efficient processes. Over the past 40 years heat exchanger network (HEN) synthesis has been seen as an important research topic in order to maximise energy recovery and minimise external energy inputs from combustion sources and cooling utilities within processes.

HEN synthesis approaches can be split into 2 rough categories, sequential and simultaneous strategies. Sequential synthesis strategies involve decomposing the problem into a number of sub-problems, usually through the use of temperature partitioning. A set of targets is then usually set that either involves minimising utility costs, number of heat exchanger units, or the heat transfer area based on the thermodynamics of the system in question. Pinch Technology is the more well-known of these sequential strategies and uses a minimum approach temperature to locate bottlenecks for energy saving known as pinch points (Linnhoff & Hindmarsh, 1983). The HEN is then designed around sub-networks above and below these pinch points in order to maximise energy recovery. Mathematical programming has also been employed in sequential synthesis solving the sub-problems mentioned sequentially. Examples of these approaches include the transhipment and transportation

mathematical programming models of Papoulias and Grossmann (1983) and Cerda, et al. (1983) respectively.

Simultaneous HEN synthesis makes use of mathematical programming and advanced mathematical solvers to optimise an objective function that can consider many different competing variables simultaneously. Yee and Grossmann (1990) presented a superstructure-based approach that makes use of mixed-integer nonlinear programming (MINLP) to minimise an economic objective function that considers costs associated with fixed costs, area costs, and utility costs. The superstructure is presented in Figure 1. This formulation allows streams to split and assumes isothermal mixing of streams in order for the feasible solution region to be linearly constrained. The model considers constant heat capacity flow-rates and constant film heat transfer coefficients. Yee and Grossmann's formulation is non-convex due to the concave area cost function and heat transfer equation and can therefore often get trapped in local optima, resulting in suboptimal networks.

Zamora and Grossmann (1998), Daichendt and Grossmann (1994), Bjork and Westerlund (2002) used global optimisation techniques to solve the problem, however due to the complexity of the problem, shortcut models are used for the individual heat exchanger designs that fail to take into account all of the factors that could impact the final cost of the network, thus limiting the usefulness of these approaches. Interestingly, one of the models of Bjork and Westerlund (2002) removed the isothermal mixing assumption, however it was found to take more time to solve and was subsequently shown to be unable to find globally optimal solutions for large problems (Huang, et al., 2012).

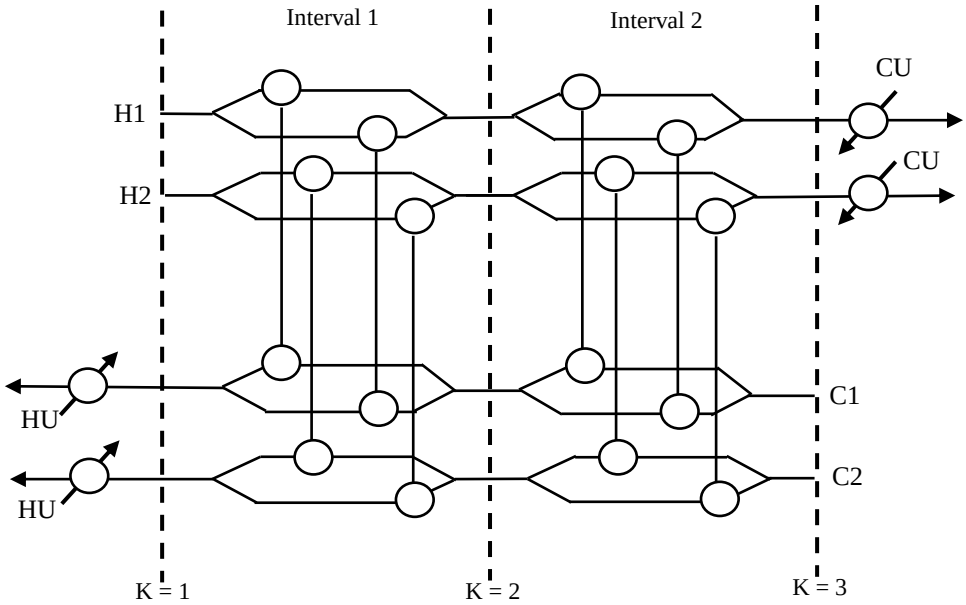


Figure 4.1: *Example of an SWS for two hot and two cold process streams and utilities as proposed by Yee and Grossman (1990).*

Huang, et al. (2012) used a novel approach for improving the SWS model (Yee & Grossmann, 1990) by adding non-isothermal mixing. The paper uses Hasan, et al. (2009) as the model's basis providing additional constraints that reduce redundant combinations in the superstructure to reduce computational complexity. These constraints use additional binary variables to ensure stream bypasses can occur and are shown to be effective in improving the solution efficiency and quality. Further variables and constraints are added to calculate the flows in split streams in order for non-isothermal mixing to take place. While Bjork and Westerlund (2002) used similar formulations, they did not propose bounds on these additional variables which the Huang, et al. (2012) do to dramatically improve the model's robustness. The effective strategies employed show the strength of the method however the authors highlight the numerical difficulties that the model still poses. The authors could not use DICOPT/GAMS due to the non-convex nature of the problem and lack of initial conditions and used BARON/GAMS with limits set on the solution times due to the slow convergence times.

Huang and Karimi (2013) presented a novel superstructure that was a stage-wise approach which allows for cross-flows, cyclic matching, matches in series on sub-streams in each stage, and multiple utilities that can be placed at any stage within the superstructure. The superstructure is larger and more complicated than that used by other authors and is capable of producing many configurations omitted by other models. The drawbacks of this are that DICOPT/GAMS is unable to solve the problem and BARON/GAMS is able to solve only after a maximum solution time is set (48 hours of CPU time in this paper). This superstructure approach was shown to be very effective for generating optimal configurations and the model expands greatly on previous works, using the Huang et al. (2012) model as its basis.

Escobar and Trierweiler (2013) compared HENS strategies and suggested systematic initialisation strategies for the MINLP SYNHEAT formulation. The systematic comparison found that the SYNHEAT model provided better solutions than sequential approaches and that SBB/GAMS and DICOPT/GAMS were the best performing solvers. (Escobar and Trierweiler, 2013)

All models reviewed thus far improve heat exchanger network synthesis by improving or expanding on these simultaneous optimisation approaches using shortcut models for individual exchangers that only take into account an approximated area for capital costing. If the networks obtained from these simultaneous approaches are designed using more detailed models for the individual exchangers, the cost of the resulting network can be

drastically different as the areas given by the approximate models are often far from the areas of the real exchangers, especially when considering stream properties and the number of shells required for a particular heat exchanger.

There have been many recent publications that use different approaches for optimising or improving individual heat exchangers, without considering network design. These include Vengateson (2010), Sanaye and Hajabdollahi (2010), Wang et al. (2012), Onishi et al. (2013), and Fettaka et al. (2013). While these studies have their individual merits, this work attempts to find a way to include detailed design considerations at the level of heat exchanger network synthesis. Relatively few attempts have been made to include design features that have an effect on the overall cost of the network, such as pressure drops, differences in overall heat transfer coefficient resulting from stream properties, and the effect of multiple shell and tube passes, into the overall network synthesis.

4.2 Combining Exchanger Design with Network Synthesis

The inclusion of exchanger design in network synthesis is of importance for obtaining a truly useful design for implementation, as many exchangers are made of special materials, different pressure ratings and varying heat transfer ratings. The Kern (1950) correlation for shell-side flow and the Dittus-Boelter correlation for tube-side flow were used by Polley, et al. (1990) to develop a relationship between pressure drop, heat transfer coefficient and exchanger area. This allowed for the possibility of including pressure drop into simultaneous network synthesis. Frausto-Hernandez, et al. (2003) implemented this in a modified version of Yee and Grossmann's (1990) SYNHEAT model, specifying maximum pressure drops and allowing for varying heat transfer coefficients related to area and pressure drop. The model showed the potential of including pressure drop in HEN synthesis however the inclusion of additional non-convex constraints resulted in numerical disadvantages when solving the problem. Their model also assumed that all exchangers were counter-flow single-pass shell single-pass tube exchangers.

Soršak and Kravanja (2002) performed a SWS superstructure optimisation with modifications that allow for the simultaneous consideration of multiple exchanger types. These modifications take the form of disjunctions at each possible exchanger match within

the superstructure as well as additional constraints that ensure feasible temperature distributions are valid for all exchanger types. Due to these additional complexities a specially developed MINLP solution approach is used where an initialisation scheme is used that halves the time it takes to solve the mixed-integer linear program (MILP) master problem in the modified outer approximation/equality relaxation (OA/ER) algorithm. The model utilised shortcut models for detailed designing of individual heat exchangers, and different cost functions for evaluating the different exchanger types and their effects on the objective function (Soršak & Kravanja 2002).

Mizutani, et al. (2003a) used a Generalised Disjunctive Programming (GDP) reformulated as an MINLP to optimise individual exchangers using the Bell-Delaware method for shell-side calculations and then extended this model to include network optimisation (Mizutani, et al., 2003b). This model makes use of disjunctions in the topology selection and at the level of individual exchanger design. The model attempts to ensure that the overall heat transfer coefficients are valid for each exchanger and iterates back to the network design to see whether the new heat transfer coefficients from the exchanger designs change the optimal network and uses the logic-based outer approximation method to do the optimisation (Turkay & Grossmann, 1996). The individual exchangers are designed as single tube- and shell-pass exchangers and are not designed to TEMA standards. This results in unrealistic exchangers with far lower pressure drops than could be expected with multiple pass exchangers. (Mizutani, et al., 2003b)

Ravagnani and Caballero (2007a) followed a similar procedure to that of Mizutani, et al. (2003a) for the syntheses of individual exchangers. However they used a tube counting table to follow TEMA standards as well as including the possibility of multiple shells and tube passes. They extended their model to include a bi-level decomposition where a SYNHEAT model is utilised during the network optimisation, followed by individual unit optimisation (Ravagnani & Caballero, 2007b). The calculated heat transfer coefficients from the individual heat exchanger units are then substituted back into the SYNHEAT model, which is subsequently re-run. This is done until 2 consecutive solutions are worse than the previous structure. This iterative procedure does not necessarily provide good network optimisation as the only parameter that is “checked” by the detailed heat exchanger optimisation is the heat transfer co-efficient. Should a significant change in heat transfer co-efficient result from a certain heat transfer match then the subsequent network optimisation can exclude that specific match, even if the heat load and transfer area are different in the subsequent topologies. While this model can obtain networks that can be reproduced in reality, it fails

to account for the costs associated with the detailed designs, such as pressure drops, number of shells, etc., in the network topology optimisation.

As far as the authors of this study are aware, there have been no further attempts to combine both grassroots network design via simultaneous optimisation strategy and detailed heat exchanger design, apart from those of Mizutani, et al., (2003b) and Ravagnani and Caballero (2007b).

It has been shown that it is impossible to solve the MINLP approaches described above to a guaranteed global optimum due to the non-convexities and combinatorial nature of the problem and it was suggested that simplified models need to be developed in order to have solutions that run in polynomial time (Furman & Sahinidis, 2002). The model presented in this work, while not improving on the network generation section in terms of offering a solution to the global optimisation of the problem, does circumvent 2 of the key problems associated with the models presented by Mizutani, et al. (2003b) and Ravagnani and Caballero (2007b). The first problem of the aforementioned models is that the individual heat exchangers are optimised using large MINLP models with discrete variables running into the thousands. The result of this is that the chances of sub-optimal individual exchanger designs could be very high, especially without a rigorous bounding strategy. By bypassing this individual heat exchanger optimisation approach and replacing it with a design based on heuristics (as done in this paper), or with another type of design approach such as using the software HTRI ™, it is possible to find a very good solution. The second problem associated with the aforementioned approaches of incorporating detailed exchanger design into network synthesis, is that the only link between the MINLP network synthesis and the individual exchanger designs was the overall heat transfer co-efficient. By providing the MINLP network optimisation with more details of the individual exchangers, such as number of shells, pressure drops, etc. it should be possible to obtain better network design.

The procedure for the synthesis of heat exchanger networks presented in this paper uses a 2-step design approach where the first step constitutes network synthesis and the second constitutes detailed individual exchanger designs. The network synthesis uses the SYNHEAT model, as discussed in detail in Yee and Grossmann (1990), which makes use of shortcut models for the individual exchangers. This model is used to avoid a highly non-linear solution space for the MINLP solvers in order to more easily find near-optimal solutions at the network synthesis level. The network obtained from the SYNHEAT optimisation is then designed in detail, to TEMA standards. The detailed designs are then used to modify the objective function of the network synthesis model in order to provide it with more

information regarding the behaviour of the actual designs on the objective function, F_T correction factors, numbers of shell and tube passes, overdesign and TEMA selections, pressure drops, and overall heat transfer coefficients. It is thus possible to exclude networks that will no longer be optimal once all of the detailed exchanger design considerations are taken into account and keep the MINLP model simple enough to consider large problems. An iterative procedure is also outlined that limits the change of each parameter between iterations so that potential solutions are not excluded. All additions to the objective function, including pressure drops and correction factors, are linear and thus do not add to the non-convexity of the objective function. The method is detailed below.

4.3 Methodology

This section will detail the methodology used in the heat exchanger network synthesis examples.

The methodology for the topology optimisation is the same as that of the SYNHEAT method presented by Yee and Grossmann (1990) with minor adjustments, so only the modifications will be presented in this paper and the reader is encouraged to refer to the original paper for the full formulation. The SYNHEAT model equations are also presented in Appendix 4.A. The main difference in the model formulation of this paper and the original SYNHEAT model is the inclusion of correction factors in the objective function that allow for it to converge to the same total annual cost as the network that is rigorously designed for after the optimisation.

$$
\begin{aligned}
min \Bigg[& \sum_{i \in H} CUC\ qc_i + \sum_{i \in C} HUC\ qh_j + CF \left(\sum_{i \in H} \sum_{j \in C} \sum_{k \in K} z_{i,j,k} NSP_{i,j,k} + \sum_{i \in H} zcu_i + \sum_{j \in C} zhu_j \right) \\
& + \sum_{i \in H} \sum_{j \in C} \sum_{k \in K} NSP_{i,j,k} AC \left(CorF_{i,j,k} \frac{q_{i,j,k}}{NSP_{i,j,k}(U_{i,j,k})(LMTD_{i,j,k})} \right)^{AE} \\
& + \sum_{i \in H} AC \left(\frac{qc_i}{(U_i)(LMTD_i)} \right)^{AE} + \sum_{j \in C} AC \left(\frac{qh_j}{(U_j)(LMTD_j)} \right)^{AE} \\
& + \sum_{i \in H} \sum_{j \in C} \sum_{k \in K} NSP_{i,j,k}.PC.z_{i,j,k} \left(delPc_{j,k} VolFc_j + delPh_{i,k} VolFh_i \right) \Bigg]
\end{aligned}
\tag{4.1}
$$

Where *CUC* and *HUC* are the costs of the cold and hot utilities respectively, $q_{i,j,k}$ is the energy transferred between hot process stream *i* and cold process stream *j* in interval *k*, qh_j and qc_i is the energy transferred from hot utility to cold stream *i* and from cold utility to hot stream *j* respectively. $z_{i,j,k}$ is the binary variable representing a process stream match between hot process stream *i* and cold process stream *j* in interval *k*. *CF* is the fixed cost associated with an exchanger, *AC* is a variable cost factor based on the area, *AE* is the area annualising factor, and *PC* is the cost associated with pumping. $NSP_{i,j,k}$ is a correction factor that accounts for the number of shell passes, discussed below. $delPc_{j,k}$ and $delPh_{i,k}$ are the pressure drops per shell pass of the cold and hot streams respectively in interval *k*. $VolFc_j$ and $VolFh_i$ are the volumetric flow-rates of the cold and hot streams respectively, inputted as parameters. $U_{i,j,k}$ are the overall heat transfer coefficients that are match-dependent and corrected for in each iteration. *LMTD* is the log mean temperature difference, calculated using the Chen (1987) approximation to avoid numerical difficulties.

$CorF_{i,j,k}$ are match-specific correction factors that are applied to the areas to make them converge on an area that can be designed outside of the MINLP formulation. These factors take the form of parameters in the model and are derived by dividing the area of the detailed exchanger design by the area obtained in the MINLP network synthesis model. These factors are correct for the non-approximated LMTD, the F_T correction factor, and TEMA decisions. This new objective function aims at encompassing all of the cost features of a network generated in a rigorous way in the topology selection, unlike those of previous authors. The cost features that are impossible to model in the MINLP network formulation due to the nonlinearities involved in the calculations of these factors are lumped and added into the objective function.

The differences between this new formulation and the objective function used in the original SWS model and the models of Mizutani, et al. (2003b) and Ravagnani and Caballero (2007b) are outlined below. Firstly, the pressure drop is now included as an implicitly calculated factor, as shown above. It is implicit as the pressure drop for a particular stream match is set for each network's generation iteration and is merely selected depending on the binary variable selected. This means that the model can choose to exclude matches with excessive pressure drops but has the weakness of not being able to design individual exchangers that can mitigate this pressure drop. As a result of this formulation stream splitting can no longer be included, unless the flow splits can be explicitly calculated in the model and included in the objective function. Notice also that the pressure drops are associated with a stream and not the shell- or tube-side, allowing for the exclusion of the highly non-linear equations

usually used in the calculation of shell-side and tube-side pressure drops in the network generation. The addition of pressure drops into the objective function as a linear implicit equation does not add to the model's solving complexity, unlike the formulation of Frausto-Hernandez, et al. (2003), in which considerable non-convex constraints were needed in order to incorporate pressure drop.

The fixed cost, *CF*, associated with each selected exchanger is now multiplied by the number of shells that are required for each match. This is done to penalise the selection of matches that would require the purchasing of multiple identical shells while at the same time accurately depicting the costs associated with the purchasing of multiple shells. This is also added into the variable area cost term by dividing the area by the number of shells that would be required for the individual exchangers and multiplying that area by the number of shells that would be needed in the series shell exchangers. Note that this would also add to the overall cost of the model and will therefore make the solutions obtained in this methodology have a higher TAC than other methods in literature.

The only further addition to the SYNHEAT model of Yee and Grossmann (1990) is the exclusion of stream splitting in order to accommodate the pressure drops as explained. The simple constraint used is:

$$\sum_{j \epsilon C} z_{i,j,k} \leq 1 \quad j \epsilon C, k \epsilon K \tag{4.2}$$

$$\sum_{i \epsilon H} z_{i,j,k} \leq 1 \quad i \epsilon H, k \epsilon K \tag{4.3}$$

Table 4.1: *Correction factors added to the SYNHEAT model to correct the objective function*

Correction factor	*Purpose*
$CorF_{i,j,k}$	Factor used to get area to converge on detailed design area
$delPc_{j,k}$	Pressure drop of cold stream (in Pa) inputted from external individual heat exchanger design
$delPh_{i,k}$	Pressure drop of hot stream (in Pa) inputted from external individual heat exchanger design
$NSP_{i,j,k}$	The number of shell passes for that particular exchanger
$U_{i,j,k}$	The overall heat transfer coefficient of the selected exchanger (W/m^2K)

After the initial SYNHEAT model is solved in GAMS with the additions mentioned above, the network is modelled using heuristics in an Excel spreadsheet using the Bell-Delaware method for shell-side calculations as well as heuristics described by Serth (2007). Note that the exchangers are therefore not rigorously optimised but rather that practical

considerations and empirical data are used. As this step is performed outside of the topology optimisation routine, other design methodologies can be used in this step such as a rigorous optimisation as performed by Ravagnani and Caballero (2003a) or to include detailed design using AspenTech's HTRI ™ software. Whichever is used, it should not have an effect on the convergence of the methodology but can result in different designs. The heuristic method was chosen for this study in order to avoid the local optima experienced in large non-convex problems such as by Mizutani, et al. (2003a) and Ravagnani and Caballero (200a7). The key focus of this paper is not the individual unit designs but rather how these designs can be used to generate multiple networks and to "guide" the MINLP model towards more realistic and yet cost effective solutions. The details of the method used for individual heat exchanger designs are included in Appendix 4.C.

Once the detailed designs are obtained, the correction factors are then calculated by dividing the area of the detailed exchanger by the area obtained in the MINLP. These factors, detailed above and shown in Table 4.1, are limited to a partial changing of some numbers to avoid the solution space being too drastically altered and potentially good solutions excluded. In the examples detailed below, the change was restricted to 10% for each of the factors. For example, if the first run of the MINLP had an overall heat transfer coefficient (U) of 750 W/m^{2}K, and the first design predicts a U of 450 W/m^{2}K for stream match (1, 1, 1), i.e. match between hot stream 1 and cold stream 1 in interval 1, then in the subsequent run the $U_{1,1,1}$ is changed to 675 W/m^{2}K (a decrease of 10% and a correction factor of 0.9). Note that this would mean that the shell pass correction factor would have non-integer values in the iterations however they will converge upon integer values by the end of the algorithm. The initial values for the correction factors should be carefully chosen so that no potential solutions are excluded. The best way for this to be done is to pick values that will underestimate the objective function (i.e. high overall heat transfer coefficients and low pressure drops). These correction factors are now inputted as parameters into the MINLP model and the model is re-solved. If the generated network is identical to the previous network and the new correction factors within a certain tolerance of each other, the convergence termination criterion is met and the procedure is stopped. If the network is not the same as the MINLP network, this network is designed using the heuristic procedure again and new correction factors are derived at. The iterative algorithm is shown graphically in Figure 4.2.

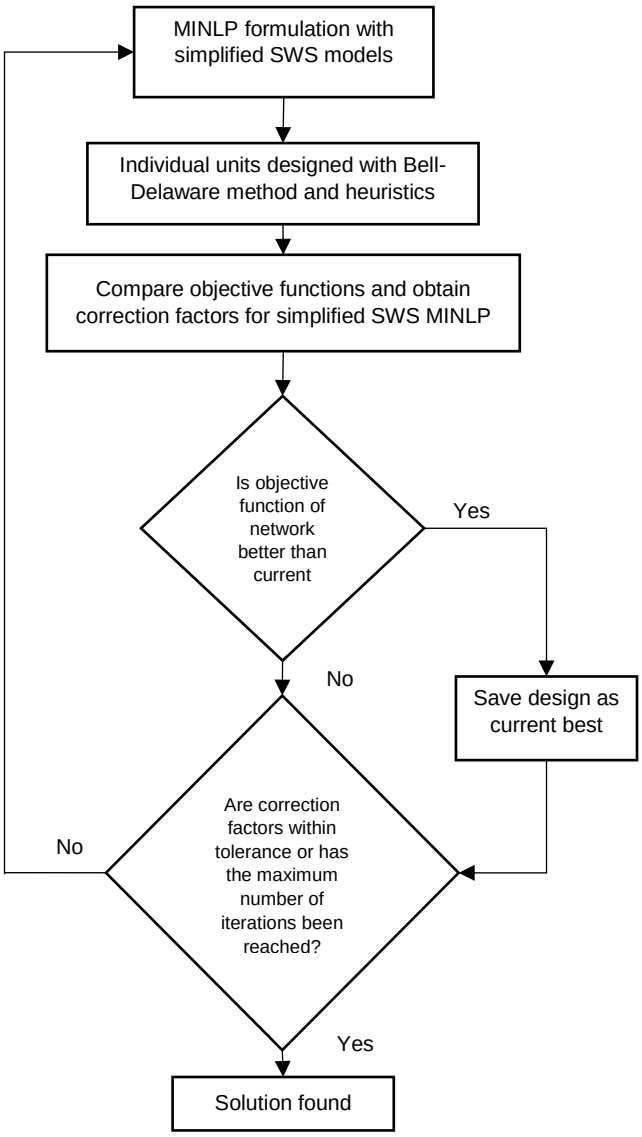


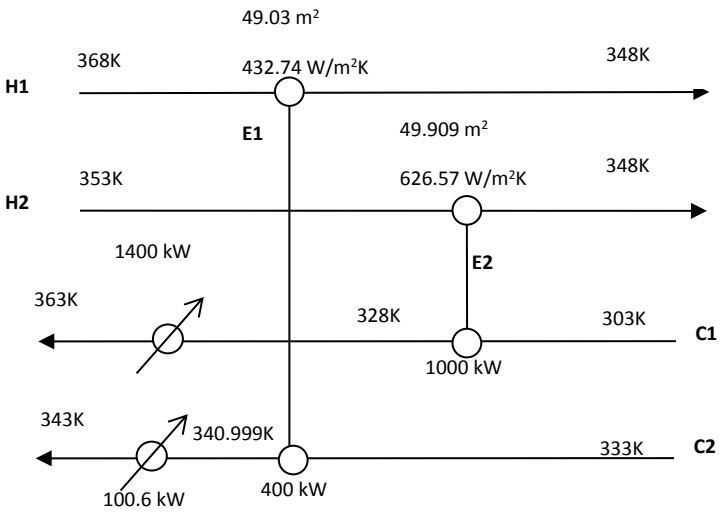
Figure 4.2: *The proposed iterative procedure used in this study.*

All these factors must be within 1 % of the solution generated in the detailed design section for the selected exchangers for the termination criteria to be met. Once the correction factors have converged so that the objective function of the modified SYNHEAT model is the same as that of the rigorously designed network, then the optimal network is selected. Convergence cannot be guaranteed, so if the maximum number of iterations is reached then this is also a termination criteria. However, applying a partial change of correction factors seems to increase the likelihood of reaching the convergence. In the examples studied, the convergence was reached. The optimal network is then described as the network having the lowest total annual cost (TAC) of all the networks generated. While this is not a rigorously determined optimum and it cannot be proved to be the global optimum, it can be shown to result in highly competitive solutions. This is mostly due to the fact that the correction

factors in the objective function progressively improve obtaining the appropriate trade-off between investment and utility cost, allowing for subsequent runs to find new networks that may be more optimal once the unit designs are taken into consideration.

4.4 Solution Algorithm

The SYNHEAT model (Appendix A) with the above changes is solved using DICOPT/GAMS with GAMS version 24.2.3 and using CPLEX as the MILP solver, CONOPT as the NLP solver and DICOPT as the MINLP solver. The computing platform had an Intel ® Core ™ i7-4700MQ 2.4 GHz CPU and 16 GB of RAM. All SYNHEAT models were solved over a few seconds and 5 minutes of CPU time.



Area cost ($/year) = 5,632, pumping cost ($/year) = 1,492, utility cost ($/year) = 90,036, global annual cost ($/year) = 97,159

Figure 4.3: *Optimal network for example 1*

Table 4.2: *Data for Example 1, from Mizutani, et al. (2003b)*

	m (kg/s)	T_{in} (K)	T_{out} (K)
H1	8.15	368	348
H2	81.5	353	348
C1	16.3	303	363
C2	20.4	333	343
HU		500	500
CU		300	320

Where ΔTmin is 10 K.

Area cost = 1000(number of shells) + $60.A^{0.6}$ $/year, where A = m^2.

Pumping cost = $0.7(\Delta P_t m_t/\rho_t + \Delta P_s m_s/\rho_s)$, where ΔP = Pa, m = kg/s, and ρ = kg/m^3.

CW cost = $6/kW.year. S cost = $60/kW.year.

Overall heat-transfer coefficients of process stream and utility matches = 444 $W/m^2.K$

Table 4.3: *Data for Example 2, taken from Mizutani, et al (2003b)*

	m (kg/s)	T_{in} (K)	T_{out} (K)
H1	134	413	313
H2	235	433	393
H3	12.1	483	318
H4	28.5	533	333
H5	102	553	483
H6	14.2	623	443
H7	38.9	653	433
C1	235	543	658
C2	143	403	543
C3	104	293	403
CU		293	298
HU		700	700

Where ΔTmin is 10 K.

Area cost = 1000(Number of shells) + $60.A^{0.6}$ $/year, where A = m^2.

Pumping cost = $0.7(\Delta P_t m_t/\rho_t + \Delta P_s m_s/\rho_s)$, where ΔP = Pa, m = kg/s, and ρ = kg/m^3.

CW cost = $6/kW.year. S cost = $60/kW.year.

Process-utilities overall heat-transfer coefficients = 444 $W/m^2.K$

Table 4.4: *Stream Data for all, taken from Mizutani, et al (2003b)*

$\mu(kg/m.s)$	$\rho(kg/m^3)$	$C_p(J/kg)$	$k(W/mK)$	$r_d(W/mK)$
2.4E-4	634	2454	0.114	1.7E-4

4.5 Case Studies

The methodology described above was applied to 2 case studies, both extracted from Mizutani, et al. (2003b). The stream data for both case studies is presented in Table 4.4 and in both examples the matches between process streams and utilities (i.e. utility exchangers) are ignored in terms of the iterative procedure in order to compare solutions with that of Mizutani, et al. (2003b) and Ravagnani and Caballero (2007b), both of whom made the same assumption.

4.5.1 Case Study 1

The first case study involved two hot process streams and two cold process streams and one hot and one cold utility in a two-stage superstructure. The data for this example is presented in Table 4.2, taken from Mizutani, et al. (2003b). The converged network was found after 11 iterations with a TAC of $97,159.3 and is presented in Figure 4.2. Due to the 10% maximum allowable change per iteration for each factor that is inputted from outside the optimisation loop, it took a number of iterations for the pressure drops and heat transfer coefficients to converge upon the design obtained. The detailed exchanger designs are shown in Table 4.5 with comparisons with other methods shown. The detailed cost breakdown compared to other authors is also shown in Table 4.6.

The implicit correction factors that are added into the model, as discussed in the methodology, are initialised at values that underestimate the objective function. The $NSP_{i,j,k}$ is initialised at 1 pass, $CorF_{i,j,k}$ begins at 1, $U_{i,j,k}$ at 750 W/m^{2}K, and the pressure drops, $delPc_{j,k}$ and $delPh_{i,k}$, are initialised at 10,000 Pa.

The MINLP model had 16 discrete variables, 65 constraints, and 61 continuous variables. The network was identified during the first run and did not change throughout the iterative procedure. The network topology was identical to those identified by the other authors but the exchangers differed. The solution obtained by Mizutani, et al. (2003b) had the lowest TAC since non-TEMA designs were considered and because only single-pass exchangers were considered. This will also guarantee lower pressure drops on the tube-side as the pressure

drops associated with the headers are significant. So while the solution obtained by Mizutani, et al. (2003b) is good, it is not necessarily a realistic network, especially if space considerations are taken into account.

Table 4.5: *Detailed designs for individual exchangers for example 1 with comparison of previous studies*

	This study		*Ravagnani and Caballero (2007b)*		*Mizutani et al. (2003b)*	
	E1	*E2*	*E1*	*E2*	*E1*	*E2*
Area (m^2)	49.033	49.91	36.12	62.30	33.30	56.20
Q (kW)	400	1000	400	1000	400	1000
LMTD (K)	20.42	34.03	-	-	-	-
F_t	0.931	0.981	0.931	0.981	-	-
N_{tp}	4	2	2	4	-	-
D_s (m)	0.489	0.4382	0.337	0.686	0.400	0.650
N_t	224	228	90	427	86	72
N_b	10	2	98	3	13	10
d_{ex} (mm)	19.05	19.05	19.05	19.05	25.4	50.8
d_m (mm)	14.83	14.83	17.01	17.01	21.18	46.58
pt (mm)	25.4	25.4	25.4	25.4	-	-
L (m)	3.6576	3.6576	6.71	2.438	-	-
h_s ($W/m^2\,^oC$)	1175	1478	2409.24	1461.14	-	-
h_t ($W/m^2\,^oC$)	1219.26	2501.7	2058.44	1795.72	-	-
$U_{overall}$ ($W/m^2\,^oC$)	432.74	626.57	582.796	480.8	588.00	523.00
ΔP_t (kPa)	25.218	11.78	11.85	8.829	-	-
ΔP_s (kPa)	13.85	6.523	2.758	1.495	-	-
arr	Square	Triangular	Square	Square	Square	Triangular
Hot fluid allocation	Tubes	Tubes	Tubes	Shell	Shell	Tubes

Table 4.6: *Comparison of solutions with other authors for Example 1*

	Mizutani et al. (2003b)	*Ravagnani and Caballero (2007b)*	*This paper*
	Overall		
Total annual cost ($/yr)	95,852.0	96,137.71	97,159.3
Area cost ($/yr)	5,608.0	5,675.52	5,631.68
Pumping cost ($/yr)	244.0	462.19	1,491.63
Utility cost ($/yr)	90,000.0	90,000.0	90,036.0

It is also worth noting that the 2 exchangers in the final design of this study are overdesigned by 0.9 % and 4.4 % for exchangers E1 and E2 respectively in the individual exchanger design section due to the heuristic that suggests a good design should be overdesigned. While this is not the only reason the method of this paper gives higher TACs, it is a contributing factor.

Interestingly as well is that the correction factors for the areas, $CorF_{i,j,k}$, converged on values of 1.083 and 1.064 for E1 and E2 respectively. This shows that the overdesign is not the only contributing factor to the $CorF_{i,j,k}$ and its inclusion can account for the F_T correction factor, differences between the LMTD approximation and the Chen approximation, and TEMA design decisions.

It is also worth noting the convergence properties of the algorithm for the correction factors and this is shown for one of the exchangers in Table 4.7. One property worth commenting on here is that the $U_{overall}$ and the $CorF_{i,j,k}$ are intrinsically linked. As $U_{overall}$ decreases the $CorF_{i,j,k}$ increases, or vice versa, due to the direct relationship. The limit on the amount of change per iteration is helpful in this regard so as to avoid solutions being omitted. As the $U_{overall}$ reaches its final value, the $CorF_{i,j,k}$ also settles on a value. This means that often the exchanger is under-designed at the start of the iterations and gradually becomes overdesigned before settling on the final value. In the larger example this allows for a certain amount of selective variety as overdesigned matches are excluded and many networks are evaluated. The other convergence property is that the pressure drop for the hot side, being the factor initialised furthest from the converged value, takes the longest to converge and added 4 extra iterations onto the algorithm. It may be argued that the pressure drops can potentially be excluded from the method, however in larger problems, such as Case Study 2, the pressure drops were found to affect the topology as exchangers with excessive pressure drops were not selected in subsequent iterations.

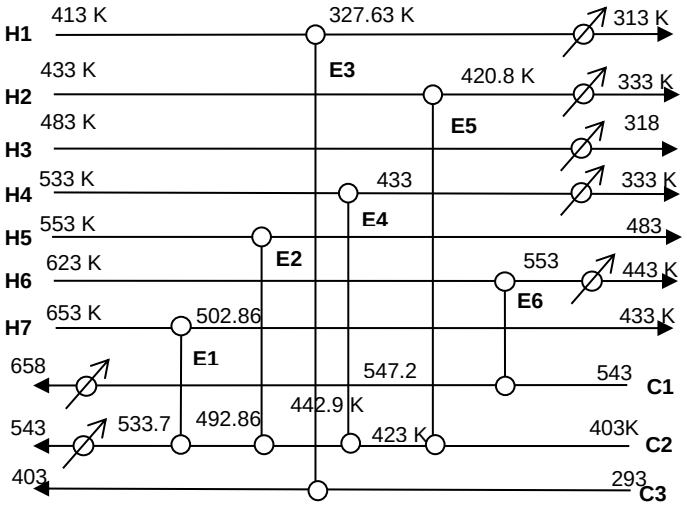
Table 4.7: *Convergence of implicit correction factors for exchanger E1*

Iteration	*CorF*	*U*	*deltaPc*	*deltaPh*
1	1.000	750.0	10000	10000
2	1.100	675.0	11000	11000
3	1.210	607.5	12100	12100
4	1.331	546.7	13310	13310
5	1.368	492.1	13850	14641
6	1.231	442.9	13850	16105
7	1.108	432.7	13850	17715
8	1.083	432.7	13850	19487
9	1.083	432.7	13850	21435
10	1.083	432.7	13850	23579
11	1.083	432.7	13850	25218

4.5.2 Case Study 2

The second case study involved seven hot process streams, three cold process streams, one hot utility and one cold utility in a six-stage superstructure. The data for this example is presented in Table 4.3, taken from Mizutani, et al. (2003b) and the stream data is the same as the previous example and is shown in Table 4.4.

This example converged to a solution after 32 iterations, producing 7 distinct network topologies during the process, with the optimal network found at iterations 5, 15, and 21 with a TAC of $4,203,057. The network that was eventually converged upon was found at iterations 2, 4, 7, 8, 9, 11, 13, and 22 until convergence at 32. The converged network is shown in Figure 4.4 and the optimal network is shown in Figure 4.5, with detailed exchanger design for the optimal network being shown in Table 4.9. A comparison between the network generated by Mizutani, et al. (2003b) and the ones obtained in this study is shown in Table 4.8, with the network of Mizutani, et al. (2003b) shown in Figure 4.7.



	Area (m^2)	$U_{overall}$ (W/m^2K)
E1	643.56	639.47
E2	626.53	662.96
E3	2553.8	700.98
E4	371.25	585.62
E5	800.06	728.82
E6	154.69	518.70

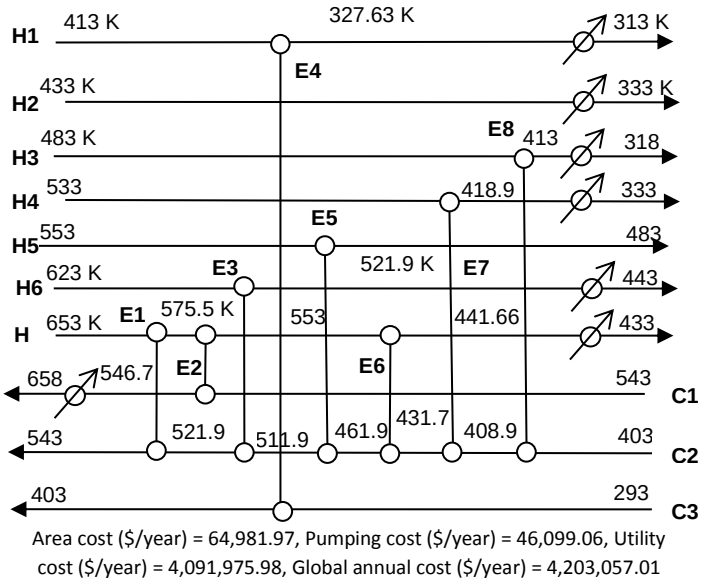
Area cost ($/year) = 67,676.734, pumping cost ($/year) = 47,592.99, utility cost ($/year) = 4,288,094.9, global annual cost ($/year) = 4,403,364.6

Figure 4.4: *Converged network from example 2*

Figure 4.6 shows the overall investment and pumping costs from the MINLP model across the procedure. It is worth commenting that the initial rise in both over the first 12 iterations was due to the correction factors slowly increasing from the initially underestimated values. Figure 4.8, showing the objective function and the costs of utilities in the objective function

does not show this same pattern. This was, in part due to the small impact of the area and pumping costs on the overall network cost as the utilities make up for over 95% of the costs in all of the solutions. The erratic nature of the objective function in this graph is explained by the 7 different topologies that were found, with different objective function values. The existence of these solutions can be explained by the possibility of different local optima within the solution space. The correction factors inserted into the model then initialise the model in different ways, thus the chance of generating different (potentially non-globally optimal) network topologies is created. This can be illustrated more obviously with the solution generated iteration 10. As the correction factors are limited to a change of only 10% per iteration and only have a minor effect on the objective function (as the utilities account for most of this), it is obvious that this solution is a suboptimal solution and thus a local optimum.

A comprehensive list of the final correction factors for all matches is shown in Table 4.A1 in Appendix 4.A.



	Q (kW)	Area (m^2)	$U_{overall}$ (W/m^2K)
E1	7398.0	148.85	666.53
E2	2148.0	177.06	754.42
E3	3523.8	210.14	539.66
E4	28074	2553.8	700.98
E5	17522	1260.9	599.19
E6	10629	544.03	639.47
E7	7978.4	426.51	550.30
E8	2078.5	130.37	588.76

***Figure 4.5**: Optimal network for example 2*

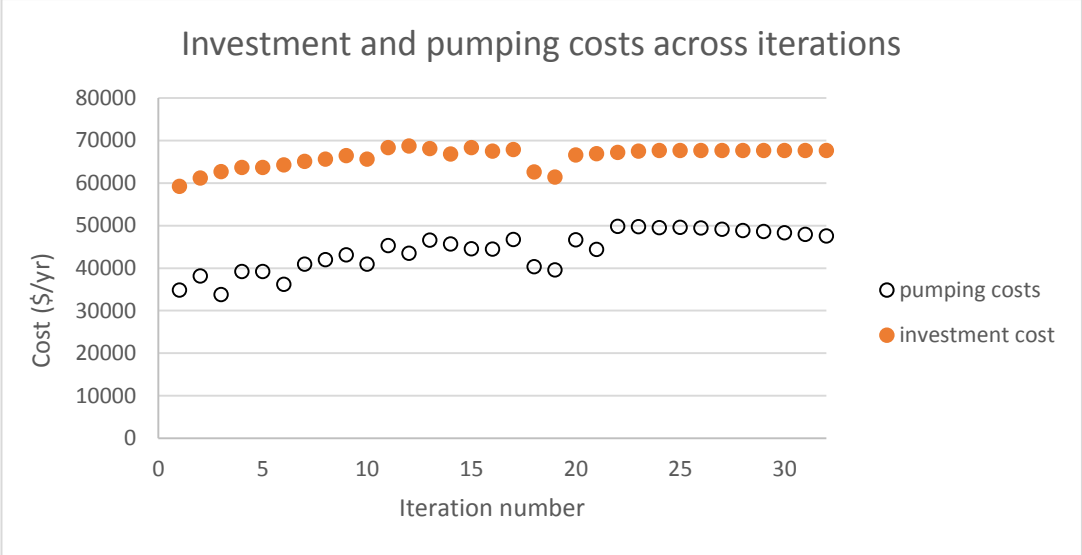
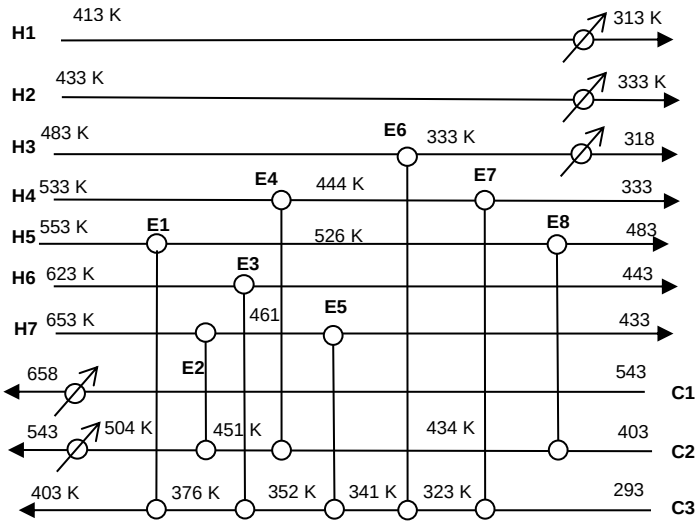


Figure 4.6: *Overall investment and pumping costs tracked over the number of iterations in the MINLP section for example 2*

Mizutani, et al.'s (2003b) solution is significantly higher than the solution obtained in this study, even though the exchangers in their paper should have no overdesign, as all variables in the model of Mizutani et al. (2003b) are continuous and not related to TEMA standards, and will have significantly lower pressure drops due to the single shell- and tube passes that were considered. Mizutani et al. (2003b) reported that the problem contained 8,452 discrete variables, 16,939 equations, and 20,408 continuous variables. It is postulated by the current authors that for this problem, the size of the feasible region resulting from the problem formulation/solution approach of Mizutani et al., (2003b) resulted in a suboptimal solution. This is due to the fact that guaranteeing a global optimum in a problem of this size is difficult, with initialisations and bounds playing significant parts in the solution generation. For comparison, the model in this paper consisted of 157 discrete variables, 629 constraints, and 542 continuous variables.



	Q (kW)	Area (m^2)	$U_{overall}$ (W/m^2K)
E1	6,80	57.8	786
E2	18,3	618	602
E3	6,30	78.2	516
E4	6,26	330	568
E5	2,72	49.5	545
E6	4,446	219	427
E7	7,74	179	594
E8	10,7	178	700

Area cost ($/year) = 24,123, Pumping cost ($/year) = 4,807, Utility cost ($/year) = 5,154,291, Global annual cost ($/year) = 5,159,098

***Figure 4.7**: Optimal network derived by Mizutani, et al. (2003b)*

***Table 4.8:** comparison of solutions with other authors for Example 2*

	Mizutani et al (2003b)	*This paper (optimal)*	*This paper (converged)*
Total annual cost ($/yr)	5,159,098	4,203,057	4,403,365
Area cost ($/yr)	24,123	64,982	67,676
Pumping cost ($/yr)	4,807	46,099	47,593
Utility cost ($/yr)	5,154,291	4,091,975	4,288,095
Number of process-process matches	8	8	6
Number of utility-process matches	5	7	7

***Table 4.9**: Detailed designs for individual exchangers for optimal solution of example 2*

	E1	*E2*	*E3*	*E4*	*E5*	*E6*	*E7*	*E8*
Area (m^2)	148.85	177.06	210.14	2553.8	1260.85	544.03	426.51	130.37
Q (W)	7398	2148	3524	28074	17521.5	10628.7	7978	2078.5
LMTD (K)	78.44	17.76	39.37	19.83	29.98	36.69	39.44	32.0
F_t	0.953	0.951	0.829	0.812	0.807	0.876	0.92	0.91
N_{sp}	1	1	1	5	2	2	2	1
N_{tp}	2	2	6	4	4	4	6	4
D_s (m)	0.635	0.635	0.787	1.143	1.372	0.9398	0.9398	0.5906
N_t	408	728	576	1050	1296	932	548	536
N_b	3	1	4	3	3	3	2	2
d_{ex} (mm)	19.05	15.87	19.05	31.75	25.4	19.05	25.4	15.87
d_m (mm)	14.83	11.66	14.83	27.53	21.18	14.83	21.18	11.66
pt (mm)	25.4	20.63	25.4	39.68	31.75	25.4	31.75	20.63
L (m)	6.096	4.876	6.096	4.876	6.096	4.876	4.876	4.876
h_s ($W/m^2\,^oC$)	1261.92	1501.33	1296.56	1195.0	926.89	1168.29	1114.48	1537.11
h_t ($W/m^2\,^oC$)	4257.27	5416.66	1901.09	6169.34	6446.12	4257.27	2324.31	2128.14
$U_{overall}$ ($W/m^2\,^oC$)	666.53	754.42	539.66	700.98	599.19	639.47	550.30	588.77
ΔP_t (kPa)	29.06	25.41	54.12	92.86	55.28	76.00	72.60	37.50
ΔP_s (kPa)	9.071	7.392	13.92	24.93	12.57	19.154	12.09	9.93
arr	Square	Square	Square	Square	Square	Square	Square	Square
Hot fluid allocation	Tube	Tube	Tube	Tube	Tube	Tube	Tube	Tube
Head Type	P	U	P	U	U	U	P	S

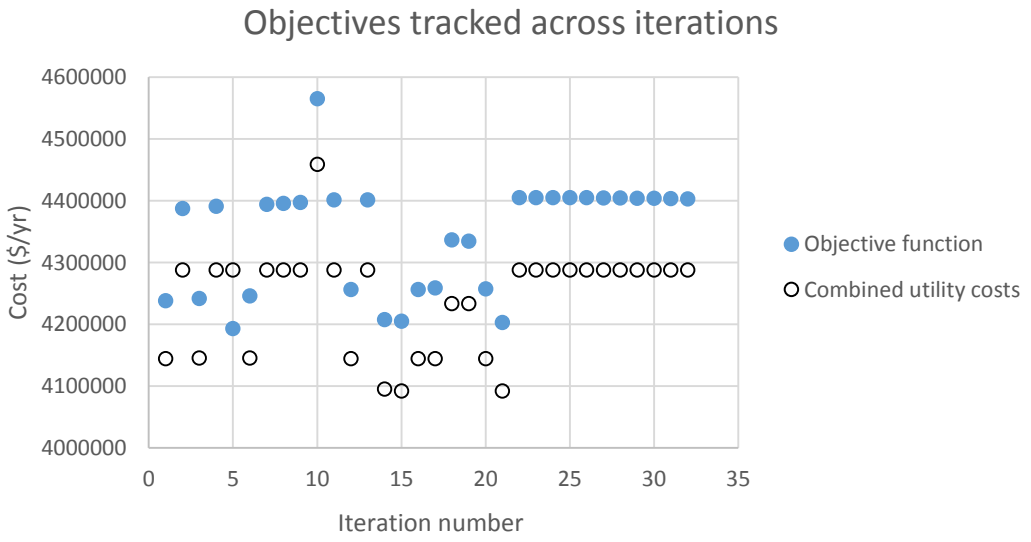


***Figure 4.8:** Objective function and combined utility costs in the MINLP section tracked across iterations for Case Study 2*

4.5.3 Comments

All of the solutions of Ravagnani and Caballero (2007b) for all examples have square tube arrangements in the reference cited. While it is possible that square arrangements are the optimal configurations in all cases, it is the suspicion of this author that this is not always the case. The choice of square arrangements in their model is more likely due to the objective function used for the individual heat exchanger optimisation subroutine that contains both pressure drop and heat transfer area considerations. The shell side pressure drop is always minimised by using square tube arrangements, where conventional optimisation literature and heuristics normally call for triangular arrangements to maximise heat transfer (Serth, 2007). It is possible that the solutions obtained by Ravagnani and Caballero (2007b) are local optima as the authors commented that the MINLP formulation is tight and needs well-formulated initialisations and bounds in order to converge on good solutions. The formulation presented in this paper therefore bypasses the issues associated with the highly non-linear nature of the problem and uses engineering judgement to seek practical solutions.

Furthermore, the optimal tube thicknesses are also not necessarily the most practical of choices, particularly for Mizutani, et al. (2003b) where large tubes are often chosen, counter-intuitively as this would decrease the contact area between the fluids.

During the iterative procedure of the second case study, it was obvious that certain matches, even when the area correction factors and overall heat transfer corrections remained constant, were excluded when pressure drops were increased. This leads to the authors of this paper concluding that it is imperative that pumping costs be included in the objective function during the topology optimisation.

While all of the problems solved here are relatively simple with regard to stream composition, it is not difficult to extend the method to include streams of varying composition. The examples here were chosen in order to compare with current literature that attempts to merge network synthesis with more detailed unit design. While this paper does not concentrate on individual design, it shows the usefulness of including detailed exchanger design considerations into network synthesis and also develops a methodology to include detailed exchanger design considerations into a network optimisation problem.

4.6 Conclusion

In this paper a methodology is developed for the syntheses of optimal heat exchanger networks, including the heuristic designs of individual exchangers. This novel methodology makes use of a two-step optimisation algorithm that uses an MINLP stage-wise superstructure approach for the initial network synthesis and a heuristic-based design approach for the individual exchangers. The objective function in the MINLP is modified in such a way as to include a variety of correction factors that allow for the simplified models in the MINLP to converge to a solution that can be simulated outside of the MINLP using more rigorous models. In the case of this paper correction factors included overall heat transfer coefficients, area correction factors (that account for over-design, TEMA standards and the F_T correction factors), pressure drops for hot and cold process streams, and the number of shell passes. The MINLP-derived at network is designed and then correction factors are inputted back into the MINLP so that the shortcut models will more closely mirror the rigorously designed exchangers. This process is repeated until the model converges on a TAC that is the same for both the rigorously designed network and the MINLP (also meaning that the correction factors are no longer changing with each successive run) or until the maximum number of iterations has been reached. The correction factors are limited in such a way that they cannot change by more than 10% with each run and all of the factors are initialised to underestimate the objective function so that potential solutions are not omitted.

In this way the proposed methodology ensures that information that is not explicitly calculated in the MINLP optimisation step is included implicitly, thereby guiding the network optimisation to solutions that will be more realistic and also excluding designs that may be suboptimal once the detailed designs are taken into account.

The methodology was applied to two case studies and successfully converged on highly competitive networks when compared to other approaches. The designs obtained follow TEMA standards and, as the pumping costs and number of shells are included in the network synthesis, are likely to give better overall solutions. The inclusion of these effects into the objective function makes comparison with other works difficult but the solutions attained are similar or better than those obtained to date.

The method of including implicit factors keeps the model's non-linearity low and allows for simultaneous consideration of factors that affect the final objective functions that have been excluded from most optimisation algorithms to date. The inclusion, in an outer loop, of a heuristic design step lowers the overall complexity of the model, therefore decreasing the chance of local optima and allowing for empirical knowledge and the designer's own experience to be included in the optimisation. For larger problems, such as the second case study, this is particularly important. The fact that the algorithm runs until the termination criterion is achieved but saves previous solutions is also important in order that an optimal solution is found and that many different networks can be assessed.

A shortcoming of the proposed approach is that the individual exchanger designs are done manually and can therefore result in suboptimal individual exchangers. This shortcoming can be remedied by using commercial heat exchanger design software, such as HTRI. In addition to this, only shell-and-tube exchangers were considered in this paper. This was done for simplicity and also in order to compare the solutions obtained to the solutions of other authors, however it is not too difficult to extend the work to include other exchangers and to apply any design method for them. A further shortcoming is that there is only an implicit pressure relationship included that is derived at externally from the individual exchanger designs. These shortcomings can be addressed in future work.

4.7 Acknowledgements

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Appendix 4A:

Table 4A.1: *Final correction factors for case study 2.*

Match	Fcor	NS	U	PC	Ph
*'1','3','3'**	1.2606	5	0.7010	4986	18571
*'2','2','4'**	1.1247	2	0.7288	10792	5538
'3','2','3'	1.0303	1.181	0.4708	4884	17716
'3','2','5'	1.1968	1.312	0.5045	6046	14641
'3','2','6'	1.0702	1	0.5888	5209	21437
*'4','2','3'**	1.1062	2	0.5856	4884	44086
'4','2','4'	1.2353	2	0.5468	10792	13310
'4','2','5'	1.1844	1.960	0.5503	6046	25939
'4','2','6'	1.2376	1.312	0.6212	5209	12072
*'5','2','2'**	1.1716	2	0.6630	8702	13993
'5','2','3'	1.2	2	0.6126	4884	16105
*'6','1','4'**	1.0496	1	0.5187	4072	54415
'6','2','2'	1.1981	1.312	0.5397	8702	16105
'7','1','2'	1.103	1.458	0.7544	7392	13310
*'7','2','1'**	1.2222	2	0.6395	11817	49751
'7','2','3'	1.1926	2	0.6400	4884	12100
'7','2','4'	1.2116	2	0.6395	10792	28533

Selected matches in the converged solution are shown by the addition of a *. All matches not shown are at initialised values, described in the text.

Appendix 4B: SYNHEAT Model Equations

The SYNHEAT model of Yee and Grossmann (1990) is presented in this section. The model equations are presented as well as the indices, sets, scalars and parameters needed for the MINLP. The only change from the formulation of Yee and Grossmann and the one presented here is the inclusion of the implicit correction factors as parameters and the modifications to the objective function discussed in the text above. The superstructure is shown graphically in Figure 4.1 in the main text.

Indices:

i	Hot process streams including utilities
j	Cold process streams including utilities
k	Interval boundary number (k = 1, ..., NOK+1)

Sets:

H	Hot process streams including utilities
C	Cold process streams including utilities
HPS	Hot process streams
HUT	Hot utilities
CPS	Cold process streams
CUT	Cold utilities
int	Intervals in the superstructure (k = 1, ..., NOK)

Scalar:

NOK	Number of intervals

Parameters:

T_i^s	Hot stream supply temperature
T_i^t	Hot stream target temperature
T_j^s	Cold stream supply temperature
T_j^t	Cold stream target temperature
F_i	Heat capacity flowrate of hot stream i
F_j	Heat capacity flowrate of hot stream j
$U_{i,j,k}$	Overall heat transfer co-efficient between hot stream i and cold stream j in interval k
CF	Fixed cost for heat exchangers
CHU_i	Cost per unit of hot utility i
CCU_j	Cost per unit of cold utility j
ACC	Area cost co-efficient
ACE	Area cost index
Ω	Upper bound for heat exchange
Γ	Upper bound for temperature difference
ε	Exchanger minimum approach temperature (EMAT)
PC	Pumping cost
$NSP_{i,j,k}$	Number of shell passes for match i,j,k

Variables:

$q_{i,j,k}$	Heat exchanged between hot stream i and cold stream j in interval k
$t_{i,k}$	Temperature of hot stream i at interval boundary k
$t_{j,k}$	Temperature of cold stream j at interval boundary k

Binary Variables:

$z_{i,j,k}$	Binary variable showing existence of match i, j in interval k
$dt_{i,j,k}$	Approach temperature between match i, j in interval k

Overall Energy Balance for Each Stream

These equations ensure that the sum of all heat exchanged by a specific stream is equal to the total heat load of the stream to attain that stream's target temperature.

$$\left(T_i^s - T_i^t\right)F_i = \sum_{k \in int}\sum_{j \in C} q_{i,j,k}\,, \qquad i \in HPS \tag{4.1}$$

$$\left(T_j^t - T_j^s\right)F_j = \sum_{k \in int}\sum_{i \in H} q_{i,j,k}\,, \qquad j \in CPS \tag{4.5}$$

Energy Balance for Each Stream over Each Interval

In addition to the overall stream energy balance, an interval energy balance is also required. Equations 4.6 and 4.7, below show the heat transferred between hot process streams, *i*, and cold process streams, *j*, in interval *k*.

$$\left(t_{i,k} - t_{i,k+1}\right)F_i = \sum_{j \in C} q_{i,j,k}\,, \qquad i \in HPS, k \in int \tag{4.6}$$

$$\left(t_{j,k} - t_{j,k+1}\right)F_j = \sum_{i \in H} q_{i,j,k}\,, \qquad j \in CPS, k \in int \tag{4.7}$$

Feasibility of Temperatures

Since all hot streams decrease in temperature from left to right within the superstructure and cold process streams increase from right to left, the following constraints are used to ensure this monotonicity of temperatures.

$$t_{i,k} \geq t_{i,k+1}, \quad i \in H, k \in int \tag{4.8}$$

$$t_{j,k} \geq t_{j,k+1}, \quad j \in C, k \in int \tag{4.9}$$

Logical Constraints

Binary variables, $z_{i,j,k}$, are set up to define the existence of a match *i,j* in interval *k*. The binary variables will have a value of '1' if the match *i,j* exists in interval *k* and '0' in the case of no match. The amount of heat transferred between the two streams in question is bound by the smaller of the heat duties of the two streams by the inclusion of Ω.

$$q_{i,j,k} - \Omega z_{i,j,k} \leq 0, \qquad i \in H, j \in C, k \in int \tag{4.10}$$

Calculation of Approach Temperatures

In order to calculate the heat exchanger area requirements of each match the approach temperatures are required. The variable, $dt_{i,j,k}$, is thus required in logical constraint equations so that the driving forces can be calculated. The binary variables, $z_{i,j,k}$, are again used to check whether a match *i,j* is present in interval *k*. If the match is absent the parameter Γ will be in use to ensure that the equation is not in use. By inclusion of this equation, negative approach temperatures are avoided, ensuring positive driving forces for heat exchanges.

$$dt_{i,j,k} \leq t_{i,k} - t_{j,k} + \Gamma\left(1 - z_{i,j,k}\right), \qquad i \in H, j \in C, k \in int \tag{4.11}$$

$$dt_{i,j,k+1} \leq t_{i,k+1} - t_{j,k+1} + \Gamma\left(1 - z_{i,j,k}\right), \qquad i \in H, j \in C, k \in int \tag{4.12}$$

Shenoy (1995) suggests setting the value of Γ to the maximum of zero so as to ensure that numerical errors do not arise due to negative approach temperatures between streams in which matches do not exist.

To avoid infinite area heat exchangers in the solutions an exchanger minimum approach temperature (EMAT) is set

$$dt_{i,j,k} \leq \varepsilon \tag{4.13}$$

Where ε is a small positive number.

Objective Function

The objective function here is the only major change to the model of Yee and Grossmann (1990). The changes are detailed in the main text.

$$\begin{aligned} min\Bigg[& \sum_{i \in H} CUC\ qc_i + \sum_{i \in C} HUC\ qh_j + CF\left(\sum_{i \in H}\sum_{j \in C}\sum_{k \in K} z_{i,j,k} NSP_{i,j,k} + \sum_{i \in H} zcu_i + \sum_{j \in C} zhu_j\right) \\ & + \sum_{i \in H}\sum_{j \in C}\sum_{k \in K} NSP_{i,j,k} AC\left(CorF_{i,j,k}\frac{q_{i,j,k}}{NSP_{i,j,k}\left(U_{i,j,k}\right)\left(LMTD_{i,j,k}\right)}\right)^{AE} \\ & + \sum_{i \in H} AC\left(\frac{qc_i}{\left(U_i\right)\left(LMTD_i\right)}\right)^{AE} + \sum_{j \in C} AC\left(\frac{qh_j}{\left(U_j\right)\left(LMTD_j\right)}\right)^{AE} \\ & + \sum_{i \in H}\sum_{j \in C}\sum_{k \in K} NSP_{i,j,k}.PC.z_{i,j,k}\left(delPc_{j,k} VolFc_j + delPh_{i,k} VolFh_i\right)\Bigg] \end{aligned}$$

Chen's 1st approximation will be used to avoid possible numerical errors in the calculation of the log mean temperature difference (LMTD) (Chen, 1987):

$$LMTD_{i,j,k} = \left[\frac{(dt_{i,j,k})(dt_{i,j,k+1})(dt_{i,j,k}+dt_{i,j,k+1})}{2}\right]^{1/3} \quad (4.14)$$

The MINLP model then minimises the TAC, Equation 4.1, in order to find the optimal network. Due to the linearity of all equations that define the feasible space, the model is solved without difficulty, however due to the non-linearity of the objective function it is still plausible that more than one local optimal solution exists due to the possibility of non-convexity (Yee & Grossman, 1990).

Constraint to avoid stream splitting

The final constraint is to ensure that stream splitting cannot occur and the pressure drop formulation presented in the objective function can be used:

$$\sum_{j\epsilon C} z_{i,j,k} \leq 1 \quad j \epsilon C, k \epsilon K \quad (4.2)$$

$$\sum_{i\epsilon H} z_{i,j,k} \leq 1 \quad i \epsilon H, k \epsilon K \quad (4.3)$$

The model presented in this paper has been solved with the solver DIPCOPT++, which uses CPLEX for the MILP and CONOPT for the NLP sub problems, all operating in the GAMS environment (Rosenthal, 2007).

Appendix 4C: Exchanger Design Equations

This appendix details the methods used to design the individual heat exchangers found as the optimal network by the MINLP model. All of the equations listed below are inputted into a Microsoft Excel spreadsheet and the variables that are chosen via heuristics are inputted in order to minimise the area and pressure drops of the exchangers and also abide by common heuristics. The methodology shown here is the one used in the paper, however any other method for detailed exchanger designs could have been used (e.g. rigorous optimisation or HTRI software). The paper's focus is on the generation of multiple alternative structures using the correction factors to guide the MINLP optimisation.

1. Inlet and outlet temperatures and the heat transferred are known from the MINLP solution. LMTD is calculated with:

$$LMTD = \frac{\Delta T_2 - \Delta T_1}{ln\left(\frac{\Delta T_2}{\Delta T_1}\right)}$$

Where ΔT_1 and ΔT_2 are the differences between the inlet temperature of the hot and outlet of the cold, and the inlet of the cold and outlet of the hot respectively

2. F_T correction factors are calculated using the Blackwell and Haydu (1981):

$$R = \frac{T_{Hin} - T_{Hout}}{T_{Cout} - T_{Cin}}$$

$$S = \frac{T_{C,out} - T_{C,in}}{T_{H,in} - T_{C,in}}$$

$$F_T = \frac{\left[\frac{\sqrt{R^2+1}}{(R-1)}\right] \log\left(\frac{1-P1}{1-RP1}\right)}{\log\left[\frac{\left(\frac{2}{P1}\right)-1-R+\sqrt{R^2+1}}{\left(\frac{2}{P1}\right)-1-R-\sqrt{R^2+1}}\right]}$$

Where

$$P_1 = \frac{1-\left[\frac{R.S-1}{(S-1)}\right]^{1/NS}}{R-\left[\frac{R.S-1}{S-1}\right]^{1/NS}}$$

Where *NS* is the number of shells

Or, if *R* = 1,

$$F_T = \frac{P_2 - \sqrt{R^2+1}/(1-P_1)}{\log\left[\frac{\left(\frac{2}{P1}\right)-1-R+\sqrt{R^2+1}}{\left(\frac{2}{P1}\right)-1-R-\sqrt{R^2+1}}\right]}$$

Where

$$P_2 = P_1/(NS - NS.S + P_1)$$

The *NS* is altered until F_T is more than 0.75 (Kern, 1950).

3. U_0 is estimated by the user and used to calculate area with the following per shell:

$$A_o\ per\ shell = q/(NS.U_o(F_T)(LMTD))$$

4. Tube size, d_o, thickness, length, L_t, pitch and configuration as well as head type are chosen using design heuristics from standard sizes according to heuristics detailed in Serth (2007).

5. These inputs can then be used to obtain an estimate for the number of tubes, $n_{t,est}$, using:

$$(A_o)_{per\ shell}/(\pi d_o L_t) = n_{t,est}$$

6. An estimate from this is obtained for the velocity through the tubes using the following:

$$a_i = \frac{\pi d_i^2}{4}\left(\frac{n_t}{N_t}\right)$$

$$v_i = \frac{\dot{V}_i}{a_i}$$

Where a_i is the overall cross-sectional area of the tubeside, d_i is the inner diameter of a single tube, v_i is the velocity of the fluid in the tube, and V_i is the volumetric flowrate of the fluid on the tubeside. N_t is the number of tube passes (either 1, 2, 4, 6, or 8) and is increased in order to increase the velocity in the tubes to a value that is low enough to prevent excessive fouling and high enough to get good convective heat transfer (usually between 1 and 2 $m.s^{-1}$ if the fluid is not water, with 1.7 $m.s^{-1}$ being close to optimal) (Serth, 2007).

7. This estimate is then used to determine the actual number of tubes, selected from the appropriate TEMA standards table. (Serth, 2007). When a tube number is

chosen, it is substituted into the equations above to give an actual area to the exchanger, and the velocity is checked to see whether it is an appropriate choice. The shell diameter (D_S) is thus known from the TEMA standards.

8. A baffle cut is now chosen, with 25% being the baffle cut used in this example for simplicity and due to its common use in industrial applications and lower impact on pressure drop (H&C Heat Transfer Solutions, 2014). The number of baffles, n_b, is then chosen such that the baffle spacing (L_b), spacing ratio and the shell-side velocities (v_s) are all within the heuristic limits, using the following:

   N_b should be such that (L_B is baffle spacing $L_B = \frac{L_t}{n_B+1}$)

   L_B> 50mm and

   $L_B > 0.2\ D_s$

   L_B < ½ X(max unsupported tube length)

   Shell side velocity (v_o) is, for

   Liquids 0.3-1.0 m/s

   $$v_o = \frac{\dot{V}_o}{a_o}$$

   where $a_o = \frac{(P_t - d_o)(D_s)L_B}{P_t}$

9. The inside heat transfer coefficient (h_i) is now calculated for the chosen exchanger using the modified Dittus-Boelter equation:

$$Nu_i = \frac{h_i d_i}{k_i} = 0.023 Re_i^{0.8} Pr_i^n \left(\frac{\mu_i}{(\mu_w)_i}\right)^{0.14}$$

   Where n=0.3 for fluid being cooled and n=0.4 for fluid being heated. μ_i is considered equal to $(\mu_w)_i$ in the examples considered where $(\mu_w)_i$ is the fluid viscosity at the wall.

$$Re = \frac{\rho v D}{\mu}$$

$$Pr = \frac{\mu C_p}{k}$$

$$Nu = \frac{hD}{k}$$

10. Using d_o & P_t, ascertain the equivalent diameter (d_e) for the shell-side

$$d_e = \frac{4\left[S_L S_T - \frac{\pi d_o^2}{4}\right]}{\pi d_o}$$

Where $S_L=S_T=P_t$ for square pitch and $S_L= 0.866P_t$, $S_T=P_T$ for equilateral triangular pitch.

And then derive at the outside heat transfer coefficient using

$$Nu_o = h_o \frac{de}{K_o} = J_h Re_o (Pr_o)^{\frac{1}{3}} \left(\frac{\mu_o}{(\mu_w)_o}\right)^{0.14}$$

J_h is the heat transfer correction factor from the Bell-Delaware method and is found graphically as prescribed by the Bell-Delaware method (Serth, 2007)

11. Using the h_o and h_i, fouling factors and tube diameters, the overall heat transfer coefficient is found using:

$$\frac{1}{(U_o)_{actual}} = \left(\frac{d_o}{d_i}\right)\left(\frac{1}{h_i}\right) + \frac{d_o \ln\left(\frac{d_o}{d_i}\right)}{2k} + \frac{1}{h_o} + \frac{d_o}{d_i} R_i + R_o$$

12. The actual required area of the exchanger can now be found:

$$(A_o)_{required} = \frac{q}{(U_o)_{actual}(F)(LMTD)}$$

The design can now be assessed as to whether it is over or under-designed. If the $(A_o)_{required}$ is larger than $(A_o)_{actual}$ then the exchanger is under-designed and the design needs to be changed. If it is overdesigned between 0 and 10 % then the design is suitable and if it is over-designed by over 10% then the design needs to be changed.

13. Once the exchanger is suitably sized, pressure drops can be calculated for the tube-side:

$$(\Delta P)_i = 2\rho_i v_i^2 N_t N_s \left[f_f \frac{L_t}{d_i} + 1\right]$$

Where f_f is calculated using the Haaland equation (Serth, 2007):

$$\frac{1}{f_f^{\frac{1}{2}}} = -3.6 \log_{10}\left[\left(\frac{6.9}{Re_i}\right) + \left(\frac{e}{3.7} d_i\right)^{\frac{10}{9}}\right]$$

Where e is the tube roughness.

As well as for the shell-side:

$$(\Delta P)_o = 8 j_f \left(\frac{D_s}{d_e}\right)\left(\frac{L_t}{L_B}\right)\left(\frac{\rho_o v_o^2}{2}\right)(N_s)$$

Where j_f is shown graphically in accordance with the Bell-Delaware method.

14. When all of these equations are inputted into the spreadsheet, the design task is made much faster and the user is able to make changes to the design and see the outputs. An experienced designer with good knowledge of the fluids, space and pumping requirements, and limitations of the specific process will be able to use these equations combined with heuristics in order to design near-optimal designs tailored by design experience.

Chapter 5

Multi-period Heat Exchanger Network Synthesis

Chapter 5: Multi-period Heat Exchanger Network Synthesis

This chapter is a verbatim reproduction of a paper published in the 105th *Applied Thermal Engineering* Journal on 17 May 2016 entitled "Two-step hybrid approach for the synthesis of multi-period heat exchanger networks with detailed exchanger design" (Short et al., 2016b). Similarly to Chapter 4, any changes in the chapter are only cosmetic to maintain consistency throughout the thesis. The chapter details the ways in which the methodology discussed in Chapter 3 is applied to the more complex example of a multi-period heat exchanger network design. The paper provides a brief summary of the relevant literature, as well as a detailed methodology section and a large case study that shows the application of the procedure and its merits to a large non-convex problem. It should be noted, with respect to the thesis in general, how the algorithm and design can easily be adapted to more complicated and very specific problems. In the case of multi-period HENS, the fact that the detailed design stage takes place with direct involvement from the designer is very important as specific adaptations to the process can be included, with appropriate correction factors included in the MINLP so that the topology optimisation has access to the updated information implicitly. This adaptability is a major strength of the approach, allowing for problem-specific adaptations to be included, with the possibility of other considerations to easily be included. The fact that the model is restarted many times with new initialisations provided by the evolving correction factors is of specific importance in this example (as described in Chapter 3), as the multi-period problem is significantly larger than the single period problem, meaning that the chances that the solver is trapped on local optima as opposed to the global optimum is greatly increased.

Two-step hybrid approach for the synthesis of multi-period heat exchanger networks with detailed exchanger design

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Abstract

In this study a novel methodology for multi-period heat exchanger network synthesis is presented. The new synthesis method aims to systematically generate many candidate networks and, through the use of more detailed individual heat exchanger designs and their evaluation over all periods, guide the network optimisation to more realistic designs. This is done by using the multi-period mixed integer non-linear programming (MINLP) stage-wise superstructure (SWS) model and modifying it to include correction factors. These correction factors enable the MINLP optimisation of the overall cost of the designed network, which uses only shortcut models of the individual exchangers, to be guided by more detailed models of the individual heat exchangers that comprise the network. The designs obtained at the topology optimisation stage thus more accurately represent an actual network. The correction factors take into account aspects of the real design, such as TEMA standards, F_T correction factors, number of shells, and changes in overall heat transfer coefficients. Each exchanger is designed to function over all periods of operation, and if this is not possible, extra exchangers are designed for the periods that cannot be satisfied. The methodology is applied to a case study that demonstrates the benefits of the proposed approach.

Keywords: multi-period, heat exchange networks, synthesis; mathematical programming

Nomenclature:

a_o	cross-sectional area of the shellside, m
a_i	overall cross-sectional area of the tubeside, m
arr	Tube arrangement
c_p	Specific heat capacity, J/(g·K)
d_{ex}	Tube external diameter, m
d_{in}	Tube internal diameter, m
D_s	Shell diameter, m
F_T	Correction factor taking multiple tube passes into account
k	Conductive heat transfer coefficient of the fluid, W/(m·K)
L_t	Tube length, m
L_b	Baffle spacing, m
n_b	Number of baffles
N_{sp}	Number of shell passes
n_t	Number of tubes
N_{tp}	Number of tube passes
Nu_i	Nusselt number
p_t	Tube pitch
q	Heat transferred, W
r_d	Fouling factor associated with fluid, W/(m·K)
ρ	Fluid density, kg/m^3
Pr	Prandtl number
Re	Reynolds number
T	Temperature, K
U_0	Overall heat transfer coefficient, W/(m^2·K)
v_i	Velocity of the fluid in the tube, m/s
v_o	Velocity of the fluid in the shell, m/s

V_i Volumetric flowrate of the fluid on the tubeside, m^3/s

$\dot{V}_o$ Volumetric flowrate of the fluid on the shellside, m^3/s

μ Fluid viscosity, kg/(m·s)

Indices:

i Hot process streams including utilities

j Cold process streams including utilities

p Period of operation

k Interval boundary number (*k* = 1, ..., NOK+1)

Sets:

HPS Hot process streams

HUT Hot utilities

CPS Cold process streams

CUT Cold utilities

int Intervals in the superstructure (*k* = 1, ..., NOK)

Parameters:

AC Area cost co-efficient, $\$/m^2$

AE Area cost exponent

AF Annualisation factor

CF Fixed cost for heat exchangers, \$/y

$CorP_{i,j,k}$ Correction parameters

CUC_j Cost per unit of cold utility *j*, \$/(W·y)

DOP_p Duration of period *p*

$F_{i,p}$ Heat capacity flow-rate of hot stream *i* in period *p*, W/K

$F_{j,p}$ Heat capacity flow-rate of hot stream *j* in period *p*, W/K

HUC_i Cost per unit of hot utility *i*, \$/(W·y)

NOK Number of intervals

$NSP_{i,j,k}$ Number of shell passes for match *i,j,k*

NOP_p Number of periods

$T_{i,p}^{s}$ Supply temperature of hot stream *i* in period *p*, K

$T_{i,p}^{t}$ Target temperature of hot stream *i* in period *p*, K

$T_{j,p}^{s}$ Supply temperature of cold stream *j* in period *p*, K

$T_{j,p}^{t}$ Target temperature of cold stream *j* in period *p*, K

$U_{i,j,k,p}$ Overall heat transfer co-efficient between hot stream *i* and cold stream *j* in interval *k* in period *p*, W/(m^2·K)

$xy_{i,j,k,p}$ Relaxed binary that determines whether an extra heat exchanger is required

$XA_{i,j,k,p}$ The area of any extra heat exchanger, m^2

$XNSP_{i,j,k,p}$ The number of shells of any extra heat exchanger that may be present

Ω Upper bound for heat exchange, W

Γ Upper bound for temperature difference, K

ε Exchanger minimum approach temperature, K

Positive variables:

$A_{i,j,k}$ Maximum area across all periods for the exchanger existing between cold process stream *j* and hot process stream *i* in interval *k*, m^2

AHU_j Maximum area across all periods for the exchanger existing between cold process stream *j* and the hot utility *I*, m^2

ACU_i Maximum area across all periods for the exchanger existing between hot process stream *i* and the cold utility *j*, m^2

$q_{i,j,k,p}$ Heat flow exchanged between hot stream *i* and cold stream *j* in interval *k* and period *p*, W

$qhu_{j,p}$ Heat flow exchanged between hot utility *i* and cold stream *j* in period *p*, W

$qcu_{i,p}$ Heat flow exchanged between cold utility *j* and hot stream *i* in period *p*, W

$t_{i,k,p}$ Temperature of hot stream *i* at interval boundary *k* and period *p*, K

$t_{j,k,p}$ Temperature of cold stream *j* at interval boundary *k* and period *p*, K

$dt_{i,j,k,p}$ Approach temperature between match *i*, *j* in interval *k* and period *p*, K

Binary variables:

$y_{i,j,k}$	Binary variable showing existence of match *i*, *j* in interval *k*
ycu_i	Binary variable showing existence of cold utility match with hot process stream *i*
yhu_j	Binary variable showing existence of hot utility match with cold process stream *j*

Abbreviations:

EMAT	Exchanger minimum approach temperature
HEN	Heat exchanger network
HENS	Heat exchanger network synthesis
IBMS	Interval based MINLP superstructure
LMTD	Log mean temperature difference
LP	Linear programming
MINLP	Mixed-integer nonlinear programming
NLP	Nonlinear programming
SWS	Stage-wise superstructure
TAC	Total annual cost
TEMA	Tubular Exchangers Manufacturers Association

5.1 Introduction

In a world increasingly aware of the effects of energy systems on the environment, and in which energy prices are unstable, ways of saving energy are vitally important. It is common practice in large chemical plants to use heat exchanger networks (HENs) as a way of reducing the need for external energy sources by maximising energy recovery from available sources within the process. Heat exchanger network synthesis (HENS) has been studied extensively since the problem was defined by Masso and Rudd (1969). The problem is not trivial as it involves the matching of multiple streams to optimise the total annual cost (TAC) of the network, comprising a trade-off between exchanger capital costing and utility costs.

An ideal heat exchanger network (HEN), while maximising profit and minimising wasted energy, should also be practical and be able to adequately handle a wide variety of operating conditions. A real plant may have variable operating conditions that vary with time; most processes are dynamic in nature with fluctuations in temperature and flowrates around a common set point, even in highly controlled circumstances. In addition to these minor fluctuations, planned changes are also possible. These may be the result of new product specifications, seasonal temperature shifts, start-up and shutdown procedures, etc. It is possible to design networks that remain operable during all of these circumstances. Verheyen and Zhang (2006) termed HENs that are operable and optimal under uncertain parameters “resilient” and those that are optimal over a certain time horizon with periodical changes “multi-period”. HENS has typically been approached in 2 distinct ways: sequential and simultaneous strategies. Sequential synthesis involves decomposing the problem into subproblems, usually through temperature partitioning. Details of these approaches can be found in Shenoy (1995) and Floudas (1995). With recent advances in computing and mathematical solvers, simultaneous approaches have become increasingly important. In these approaches it is possible to optimise an objective function while evaluating many competing variables simultaneously in a rigorous optimisation framework.

Most modern approaches for HENS are based around the superstructure-based approach proposed by Yee and Grossmann (1990), known as the SYNHEAT model. This approach formulates the problem as a mixed integer non-linear program (MINLP) with a stage-wise superstructure (SWS) that attempts to embed many possible network topologies. The SWS superstructure is shown in Figure 5.1 and does not rely on temperature or enthalpy intervals

or divisions. The advantage of this formulation is that none of the constraints are non-linear due to assumptions that include isothermal mixing, no series exchangers on split streams, utilities placed at the ends of the superstructure, and no stream bypasses are permitted. Yee and Grossmann's (1990) model contains constant heat capacity flow-rates and constant film heat transfer coefficients, as well as simple heat exchanger models that do not take into account the implication of multiple shells and F_T correction factors on the final solution. Even though the model is linearly constrained, it is non-convex due to the area cost term in the objective function. This can lead to sub-optimal results as there is no way of guaranteeing a globally optimal solution.

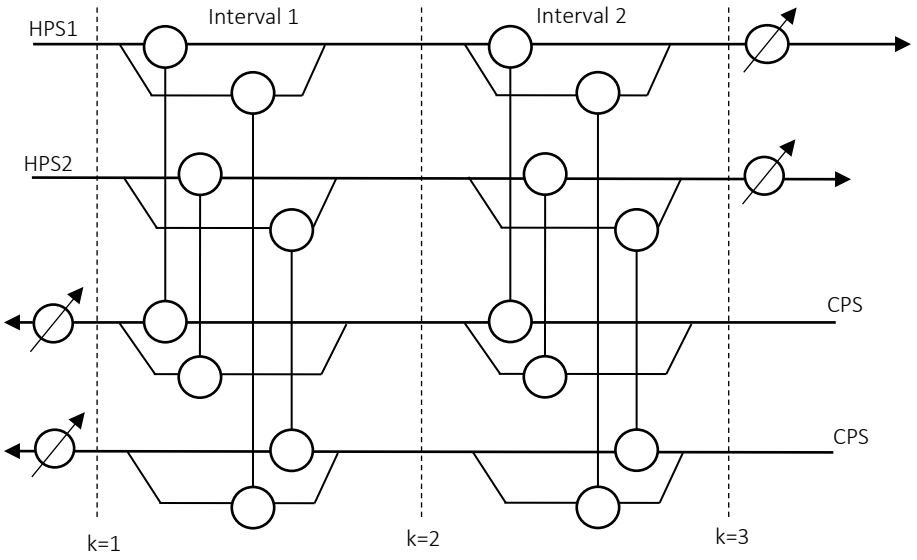

Figure 5.1: *Example of an SWS superstructure for two hot and two cold process streams as proposed by Yee and Grossmann (1990)*

This formulation has proven very popular in the process synthesis community with many subsequent papers on its improvement in terms of global solutions as well as finding uses in other applications. Escobar and Trierweiler (2013) compared a host of case studies, including methods based on the SYNHEAT model, with the goal of improving computational time and problem convergence, proposing initialisation strategies. Their study demonstrated the fact that HENS is a complex problem that, despite all of the research on the topic, still requires improvement in terms of computational efficiency. Most existing models deal with single period examples and simplified exchanger models.

Aaltola (2002) used an MINLP approach to simultaneously optimise multi-period heat exchanger networks by expanding the Yee and Grossmann's (1990) model to multiple periods. The approach used the same superstructure and many of the same constraints as

the SYNHEAT model, but included a new set that indicated the periods as well as an initial MINLP step that uses the average of the selected areas utilised over the periods. Due to the averaging of the areas over the periods, the exchanger costs are underestimated and an LP/NLP model and search algorithm is used after the network structure is found (Aaltola, 2002).

Verheyen and Zhang (2006) improved on Aaltola's (2002) model by allowing for a maximum area instead of average area to be included in the objective function. This came at a cost of adding non-linear constraints into the model. This means that, if an exchanger is active in multiple periods for the same pair of stream match in a stage, the period that results in the largest area will be the area represented in the objective function. An NLP sub-optimisation step is also included to avoid the isothermal mixing assumption. The model provided better solutions than that of Aaltola (2002). Isafiade and Fraser (2010) used an interval based MINLP superstructure (IBMS) model, where the intervals are defined by supply and target temperatures, to solve multi-period HENS problems. Using a similar formulation to that of Verheyen and Zhang (2006), they extended it to include periods of unequal durations.

Sadeli and Chang (2012) considered some of the drawbacks of MINLP approaches to multi-period HENS and developed a procedure that made use of MINLP optimisation for the initial network and then developed a set of heuristics to incorporate time-sharing. Their approach used F_T correction factors calculated in MATLAB externally once the initial MINLP was solved. These F_T factors were then inputted into an NLP sub-problem with fixed network topology. Once the network is obtained heuristics are applied to determine effective time-sharing schedules and including the addition of auxiliary exchangers where necessary (Sadeli & Chang, 2012).

Jiang and Chang (2013) decomposed the multi-period problem into multiple single period optimisations and then used a time-sharing plan similar to that of Sadeli and Chang (2012). Jiang and Chang (2015) further improved on the timesharing mechanisms by using a generic area allocation algorithm and by solving an MINLP model. Kang, et al. (2015) presented a new method for synthesis of multi-period heat exchanger networks where the authors selected a representative period which is then solved as a single period problem. The resulting process parameters and heat transfer areas in the solution network are further optimised to cater for the heat load requirements of the other periods. A second step, known as stepwise simplifying method, is then used to improve the resulting multi-period HEN of the first step through structure merging and substitution with multi-stream heat exchangers.

Kang and Liu (2014) developed a reverse order matching approach and the use of the matching algorithm of the bipartite graphs for the retrofit of multi-period heat exchanger networks. Pintarič and Kravanja (2015) developed a methodology for the synthesis and design of flexible heat exchanger networks having large numbers of uncertain parameters. Their approach entails generating an optimal heat exchanger network based on the nominal conditions, after which a flexible network is designed using reduced set of scenarios that include nominal point and critical vertices. The flexibility of the final network is then tested using a set of proposed methodologies. Li, et al., (2015) also presented a new methodology for the synthesis of flexible heat exchanger networks where for large non-convex heat exchanger problems, the degree of flexibility were established as well as the direction of deviation of uncertain parameters using direction matrix. The works of Pintarič and Kravanja (2015) and Li, et al., (2015) are focused on designing heat exchanger networks that are capable of feasibly transferring heat among streams irrespective of uncertain variability in stream parameters within a specified range. However this paper focuses on designing networks that are capable of exchanging heat among streams for cases where stream parameters vary within specified ranges.

It is worth stating at this point that when designing a network that is effective across all periods it is important to consider detailed exchanger models as the use of shortcut models cannot demonstrate whether the exchangers can perform their intended functions across all periods when fluid properties and flowrates are considered. All methods reviewed thus far have not included detailed exchanger considerations of this nature.

In order to circumvent numerical issues that result from the large sizes of these multi-period problems many decomposition approaches have been proposed, including Isafiade et al. (2015), Escobar et al. (2014), as well as stochastic methods, such as Ma et al. (2007), and Ahmad, et al. (2012). This leads the current authors to suggest that, at the present level of modern computing power and MINLP solver technology, there is little room to expand on the models currently proposed in terms of MINLP model complexity as this will lead to solutions that cannot be guaranteed to be globally optimal and can sometimes lead to poor, locally optimal solutions. Even current models that use simplified representations of the heat exchangers themselves, without considering changes in specific heat capacity (c_p), overall film heat transfer coefficients (U_0), and mostly linear constraints, may require special initialisation and bounding strategies and decomposition to find better solutions.

The lack of detailed exchanger models at the level of the network optimisation has hardly been addressed for multi-period networks; however there have been some efforts in this

regard for single period networks. Wang et al. (2012) included heat transfer enhancements in the retrofit of heat exchanger networks. Polley, et al. (1990) considered pressure drops in heat exchanger network optimisation. Soršak and Kravanja (1999) and Souza et al. (2016) also included pressure drops in detailed heat exchanger design. In the case of Soršak and Kravanja (1999), trade-offs were established between investments, utility and power consumption, while Souza et al. (2016) considered pressure drops within tubes and shells of heat exchangers as well as within connecting pipelines. Frausto-Hernandez, et al. (2003) removed the assumption of constant heat transfer coefficients and replaced it with the effect of allowable pressure drop. Soršak and Kravanja (2002) included multiple types of exchangers in heat exchanger network synthesis, Odejobi, et al. (2015) included incorporation of enhanced heat transfer techniques, while Liu et al. (2015) considered fouling, aging, and cleaning in their method. Wu, et al. (2015) developed an approach for HENS where non-isothermal mixing models and temperature-dependent heat capacities are used. In place of the conventional binary variables, the authors used a non-linear term in the objective function to convert their model into a non-linear program (NLP).

Perhaps the most comprehensive attempts at including aspects of detailed exchanger design into single period network synthesis are those of Mizutani et al. (2003b) and Ravagnani and Caballero (2007b). Mizutani et al. (2003b) in their methodology used the SYNHEAT model for the network synthesis with disjunctions that allow for the optimisation of the individual exchangers. The method used by these authors for individual exchanger optimisation is that presented previously by Mizutani et al. (2003a) where Bell-Delaware calculations are set up as an MINLP for individual exchanger synthesis of single tube- and shell-pass exchangers. The overall heat transfer coefficient (U_0) from the detailed exchanger design optimisation is then used to update the U_0 in the network optimisation. The model lacks the inclusion of the effects of multiple shells and F_T correction factor, TEMA (1998) standard choices, and pressure drops on the objective function in the network synthesis. The only extra information that the detailed designs provide to the network topology is the updated U_0. A further drawback is that, by including disjunctions at the level of both network and exchanger design, there is a high chance that locally optimal solutions will be reached, especially in large non-convex models of this type. In an example containing 7 hot streams, 3 cold streams and 1 hot and cold utility, the model contained 16,939 equations with 20,408 continuous variables and 8,452 discrete variables.

Ravagnani and Caballero (2007a) optimised individual heat exchangers using TEMA (1998) standards by implementing a tube-counting table in a large MINLP that also allowed for the

inclusion of multiple shells. In a second paper (Ravagnani & Caballero, 2007b), they extended this with a bi-level decomposition in which the SYNHEAT model is used to optimise the network topology and the individual heat exchangers are optimised using the approach of Ravagnani and Caballero (2007a). The U_0 obtained from the individual heat exchangers is then substituted back into the SYNHEAT model which is then re-run. This procedure is repeated until a subsequent run is worse than the previous run. The drawbacks of this study are the same as those of Mizutani, et al. (2003b), with an added disadvantage that the model size is much larger than that of Mizutani, et al. (2003b) with many non-linear constraints at the heat exchanger optimisation level. Difficulty in solving and finding upper and lower bounds on variables was reported and it was likely that their solutions were not globally optimal. Added to these drawbacks is that no limit was placed on the changes to U_0 between runs. This would mean that if a large change in U_0 were to occur it is likely that potential solutions will be excluded at the topology selection level.

The works of Mizutani, et al. (2003b) and Ravagnani and Caballero (2007b) are the only set of papers attempting to incorporate detailed individual exchanger designs in network topology optimisation. However, these techniques were developed for single period scenario only. The only attempt at including more detailed exchanger designs into multi-period HENS, to the best of the knowledge of the current authors, has been the work of Sadeli and Chang (2012) where F_T correction factors were included in an NLP sub-optimisation step after an MINLP optimisation step had been used to determine the network topology and intermediate temperatures. Their method also allowed for time-sharing heuristics to be applied, which gives more realistic designs that may be seen in real-life applications, however these mechanisms are only applied after the topology is fixed. Jiang and Chang (2013) used a similar time-sharing approach, however decomposed the problem into multiple single-period optimisations and then applied time-sharing mechanisms to get a combined flexible network. Neither approach includes detailed exchanger design to check whether the time-sharing will be possible when considering key design parameters such as changing U_0, F_T correction factors, number of shells, TEMA (1988) design decisions, etc.

While these studies are useful in determining individual exchanger designs in network synthesis, this work attempts to include details of individual exchanger design at the level of topology network optimisation so as to guide multi-period network synthesis to a more realistic picture of the actual network. Many studies have attempted to improve on the SYNHEAT model by adding non-convex constraints or additional binary variables to include aspects such as non-isothermal mixing, pressure drops, non-constant heat capacities,

exchanger characteristics etc., however solution times are significantly longer, feasible solutions are more challenging to attain, and global optimality is not achieved. The current study does not aim to improve these problems, however it uses a systematic approach to include more accurate data from detailed individual exchanger designs for the network topology optimisation of the multi-period system, while maintaining a relatively simple MINLP formulation with no non-convex constraints.

The methodology presented in this paper uses the MINLP formulation of Verheyen and Zhang (2006) for multi-period HENS, however with a modified objective function for the multi-period network optimisation problem. The individual exchangers in the resulting network are then designed in detail using heuristics as outlined in Serth (2007) and in Appendix A. The current authors are of the view that in a multi-period problem it is useful to use heuristics as an experienced engineer is able to test the designed exchanger over multiple periods of operation, and decide whether extra exchangers will be necessary in order for the exchanger to be used for the same pair of stream matches in all periods where the streams exist. Once the designer is satisfied with the detailed design, corrections are then computed and inputted into a subsequent MINLP network optimisation run. These corrections are inputted into the modified objective function and serve to ensure that the objective function more closely represents the objective function of the network that is designed in detail. The corrections account for the existence of extra exchangers, overall heat transfer coefficients, the existence of multiple shells, F_T correction factors, and TEMA (1988) design choices over all the periods. The subsequent run then uses this information to generate a new network which is then designed and new correction factors are obtained. This is done until either the correction factors remain unchanged over 2 consecutive runs, or until a maximum number of iterations is reached. The network that is then chosen as optimal is the one with the lowest TAC for the detailed design out of all of the designs obtained. While the methodology does not guarantee a globally optimal solution, it is successful at generating many candidate networks, guided by the detailed information provided by the exchanger heuristic design step. The methodology is detailed below.

5.2 Methodology

This section details the methodology used in solving the multi-period heat exchanger network synthesis problem in this paper and follows on from this method's application to single period use in Short, et al. (2015). The methodology section is broken into three distinct sections whereby in 2.1 the MINLP optimisation is described, the detailed exchanger designs and heuristics are briefly outlined in section 2.2, and finally in 2.3 the iterative linking process between the optimisation models and the detailed exchanger designs is explained.

5.2.1 MINLP Optimisation Model

In this paper, the SWS model of Yee and Grossmann (1990), shown in Figure 1, is extended to multiple periods of operation in a very similar way to the approach of Verheyen and Zhang (2006) and Isafiade and Fraser (2010). This is done by including a new index *p* into the model equations that accounts for the finite periods of operations, as done by Aaltola (2002). The full model, with all equality and inequality constraints, can be found in Verheyen and Zhang (2006) and Isafiade and Fraser (2010). The exchanger areas represented in the objective function are the maximum areas for each of the same pair of stream match over all periods to ensure that the exchanger is large enough to perform the required heat duty exchange in all periods, as detailed in Verheyen and Zhang (2006). The only change to the MINLP model of Verheyen and Zhang (2006) are changes to the objective function.

5.2.2 Objective Function

In order to alter the objective function so as to include information found in the detailed exchanger designs that are performed outside of the MINLP optimisation, a number of correction factors are introduced. These correction factors are implemented to get the objective function of the multi-period MINLP optimisation to more closely resemble the objective function of the detailed design performed outside of the multi-period MINLP. The correction factors are derived implicitly outside of the model by comparing the areas of the

detailed exchanger designs to the areas obtained by the shortcut models used in the multi-period MINLP optimisation step. The objective function is shown below in Equation 5.1. The objective function includes many aspects of the detailed exchanger designs, while not introducing any further model complexity in the way of non-linear constraints. In doing this the topology optimisation can be "guided" by information derived outside of the multi-period MINLP formulation from detailed individual exchanger models. Note also that this new objective function will result in higher TACs than is usually found in literature, but will incorporate many details excluded by previous synthesis models.

$$min\left\{\begin{array}{c}\left[\left(\frac{DOP_p}{\sum_{p=1}^{NOP} DOP_p}\right)\left(\sum_{i\epsilon HPS}\sum_{p\epsilon P} CUC_j \cdot qcu_{i,p}\right)\right]+\left[\left(\frac{DOP_p}{\sum_{p=1}^{NOP} DOP_p}\right)\sum_{j\epsilon CPS}\sum_{p\epsilon P} HUC_i \cdot qhu_{j,p}\right] \\ AF\left[\begin{array}{c}\sum_{i\epsilon HPS}\sum_{j\epsilon CPS}\sum_{k\epsilon K} CF \cdot NSP_{i,j,k} \cdot y_{i,j,k} + \sum_{i\epsilon HPS} CF \cdot ycu_i + \sum_{j\epsilon CPS} CF \cdot yhu_j \\ +\sum_{i\epsilon HPS}\sum_{j\epsilon CPS}\sum_{k\epsilon K}\sum_{p\epsilon P} CF \cdot XNSP_{i,j,k,p} \cdot xy_{i,j,k,p} \cdot y_{i,j,k} + \sum_{i\epsilon HPS} AC \cdot (ACU_i)^{AE} + \sum_{j\epsilon CPS} AC \cdot (AHU_j)^{AE} \\ +\sum_{i\epsilon HPS}\sum_{j\epsilon CPS}\sum_{k\epsilon K}\sum_{p\epsilon P} AC \cdot XNSP_{i,j,k,p} \cdot xy_{i,j,k,p} \cdot y_{i,j,k} \cdot \left(\frac{XA_{i,j,k,p}}{XNSP_{i,j,k,p}}\right)^{AE} \\ +\sum_{i\epsilon HPS}\sum_{j\epsilon CPS}\sum_{k\epsilon K} AC \cdot NSP_{i,j,k} \cdot \left(CorP_{i,j,k}\frac{A_{i,j,k}}{NSP_{i,j,k}}\right)^{AE}\end{array}\right]\end{array}\right\} \quad (5.1)$$

In the compound objective function (Equation 5.1) all the individual parameters and variables are defined in the Nomenclature section of this paper. Uncommon parameters, termed "corrections" in this paper, are detailed in section 5.2.3, below.

5.2.3 Correction Parameters

All corrections that are used in the model are presented in Table 1 and their implementation into the objective function is shown in Equation 5.1, above. The purpose of the correction parameters is to correct the key factors in detailed exchanger design that affect the objective function. This is done by comparing the solutions obtained by the multi-period MINLP optimisation, which uses shortcut models that do not take into account the details of actual exchanger design, to the solutions of the detailed individual exchanger designs, performed using heuristics and the Bell-Delaware method (Bell, 1963; Bell, 1981), discussed briefly in section 5.2.3 and in detail in Appendix 5A.

Table 5.1: *Corrections added into the model to correct the objective function and updated in each iteration using the detailed models.*

Correction	*Purpose*
$CorP_{i,j,k}$	Parameter used to get area to converge on detailed design area that accounts for overdesign, F_T correction factor, TEMA decisions, unapproximated LMTD etc.
$U_{i,j,k,p}$	The overall heat transfer coefficient of the selected exchanger W/(m^2·K)
$NSP_{i,j,k}$	The number of shell passes for that particular exchanger
$xy_{i,j,k,p}$	Relaxed binary correction parameter indicating the existence of an extra exchanger
$XA_{i,j,k,p}$	The area of an extra exchanger
$XNSP_{i,j,k,p}$	The number of shells required by the extra exchanger

$CorP_{i,j,k}$ corrects for the unapproximated LMTD, the F_T correction factor, and TEMA (1988) decisions. The parameter is determined by dividing the area of the detailed exchanger design for that match by the area obtained from the shortcut model in the multi-period MINLP optimisation. $U_{i,j,k,p}$ is the overall heat transfer co-efficient for a specific match in a specific period. This allows for different overall heat transfer coefficients in different periods for each match that can depend on flow-rates, heat capacities, fluid properties etc., which may change in each period of operation. Again, these are obtained from the detailed exchanger designs and inputted as parameters each time the multi-period MINLP model is re-run. $NSP_{i,j,k}$ is a parameter that is externally derived that supplies the optimisation with the information regarding the number of shell passes that a chosen exchanger may require. This is determined from the detailed exchanger designs, discussed in Section 5.3. Note the changes in the objective function formulation that is included in this paper to accommodate this. Firstly the fixed cost associated with purchasing an exchanger is now multiplied by the number of shells that will be required, not the number of matches as has been the case in previous single and multiple period studies. This is a more realistic scenario as shells in series are designed identically and each one requires its own shell and header, welding and piping, that add to the capital cost of the network. In addition, the variable cost, associated with the area, is also modified to include this. This is done by dividing the total area (of all the series shells) by the number of exchangers so that the area of a single exchanger is calculated. This is then multiplied by the number of shells.

The terms

$$\sum_{i \epsilon HPS} \sum_{j \epsilon CPS} \sum_{k \epsilon K} \sum_{p \epsilon P} CF \cdot XNSP_{i,j,k,p} \cdot xy_{i,j,k,p} \cdot y_{i,j,k}$$

and

$$\sum_{i \epsilon HPS} \sum_{j \epsilon CPS} \sum_{k \epsilon K} \sum_{p \epsilon P} AC \cdot XNSP_{i,j,k,p} \cdot xy_{i,j,k,p} \cdot y_{i,j,k} \cdot \left(\frac{XA_{i,j,k,p}}{XNSP_{i,j,k,p}} \right)^{AE}$$

found within Equation 5.1 allow for the inclusion of extra exchangers. These extra exchangers may be necessary to serve a single period or specific number of periods, if the representative match selected cannot perform the heat duty across all periods. This is likely if flow-rates are significantly different for a stream in different periods of operation as the maximum area is always chosen as the selected exchanger. A stream with a small flow-rate in a period may have vastly different heat requirements for a match, or will have a very low velocity, resulting in excessive fouling and low heat transfer coefficients. $XNSP_{i,j,k,p}$ is the parameter that translates to the number of shell passes of the extra exchanger, in the same way as $NSP_{i,j,k}$ is used, except that it is period-dependent. $xy_{i,j,k,p}$ is a binary parameter that is '1' when an extra exchanger is required and '0' when an extra exchanger is not required. Notice how this is multiplied by the binary variable $y_{i,j,k}$ in every instance of its use. This is to ensure that when a specific match is not selected, the extra exchanger that may exist for that match is also not selected. While the purpose of the parameter $xy_{i,j,k,p}$ is to take the value of '0' or '1', it is relaxed to take values between 0 and 1 in some cases. This will be further clarified in Section 5.3. $XA_{i,j,k,p}$ is another externally derived parameter that refers to the size of the extra exchanger that may be required. This exchanger area is the combined area of all shells and is analogous to the $A_{i,j,k}$. The value of $XA_{i,j,k,p}$ will always be less than that of $A_{i,j,k}$ as this is the maximum area over all of the periods and $XA_{i,j,k,p}$ refers to the area of any smaller (extra) exchanger that may be required for the specific match if the area $A_{i,j,k}$ is too large. Terms containing $xy_{i,j,k,p}$ in the objective function are initially set at zero as all of the parameters in these terms are initialised to a value of zero, meaning it is assumed that there are no extra exchangers required to carry out the heat duties throughout all periods of operation (i.e. the exchanger with the maximum area across all periods will carry out the duty of all other periods).

5.2.4 Solution Tools

The model described above, including the constraints which can be found in Verheyen and Zhang (2006), is solved using DICOPT/GAMS with GAMS version 24.2.3 and using CPLEX as the MILP solver, CONOPT as the NLP solver and DICOPT as the MINLP solver. The computing platform had an Intel ® Core ™ i7-4700MQ 2.4 GHz CPU and 16 GB of RAM. Every instance the MINLP optimisation problem was run, it solved in between a few seconds and 15 minutes of CPU time.

5.2.5 Detailed Exchanger Designs

Once a network is generated using the multi-period MINLP optimisation, this network is then designed using the approach temperatures and heat balances from the multi-period MINLP optimisation. The design can be done using any approach for the individual exchanger design, such as through the use of software packages like AspenTech's EDR ™ or HTRI ™ that make use of rigorous models, through the use of optimisation models such as that of Ravagnani and Caballero (2007a), or through the use of heuristics in an Excel spreadsheet using the Bell-Delaware (Bell, 1963; Bell, 1981) method for shell-side calculations. In the case of this study, the heuristics are used for the design of the individual exchangers and the method proposed by Serth (2007) is followed. While this means that the heat exchangers are not rigorously optimised, it has the advantage that an experienced designer with knowledge of the fluids involved can take special considerations for the fouling, fluid properties, space requirements etc., during the design process. In addition to this, during multi-period synthesis, the designer can use their engineering judgement and knowledge of the process' temperature and control requirements to decide whether the designed exchanger will be suitable to carry out the task required across all of the periods concerned. The decision to use other methods for the individual exchanger designs will have no effect on the convergence of the method. The key focus of this paper is to allow for these detailed exchanger designs to "guide" the multi-period MINLP optimisation step, not necessarily the design of individual exchangers.

5.2.6 Feasibility of Design

An important consideration in this paper is whether the designed exchanger can participate successfully in all of the periods of operation. During the design the largest exchanger for the same stream pair matches in different periods (i.e. the maximum area) is designed and then the parameters from the other periods of operation are tested to see whether this maximum area exchanger is able to perform the required duty. In this study, an overdesign of 15% for an exchanger was deemed to still be an acceptable level of overdesign (Bennet et al., 2007; Edwards, 2008). In many cases the exchanger is between 2 to 15% overdesigned, within the acceptable limits; however, if the exchanger is larger than this for a certain period then before the addition of an extra exchanger we should consider a possibility to use a portion of the shells already in place to do the heat transfer. This would therefore mean that the multi-shelled exchanger, though varying in the number of shells used per period, would be suitable across all periods and therefore there would not be a need of an extra heat exchanger for this match. This is a rare case, as it is often the case that the exchanger with fewer shells has different flowrates or F_T correction factors that would require a separate design in this period.

If an extra exchanger is required for any of the other periods, then this exchanger is also designed in the same method described in section 5.2.2. The extra exchangers will be used to obtain the correction parameters $xy_{i,j,k,p}$, $XA_{i,j,k,p}$, and $XNSP_{i,j,k,p}$. Note how these heuristics for testing the exchanger across all periods may be very difficult to automate or optimise, and hence the importance of involvement of the designer at this level of the procedure to generate feasible networks.

Sadeli and Chang (2012) and Jiang and Chang (2013) made use of time-sharing mechanisms, in order to cut down on network costs and reduce the degree of heat exchanger overdesign, by sharing exchangers that are not in use in certain periods. This may not be possible in many cases in reality, as piping and instrumentation, and the physical locations of exchangers, may make these mechanisms impossible. Added to this, exchangers may need to be cleaned to avoid undesirable mixing or contamination. Due to these reasons time-sharing was not considered in this study.

Once a feasible network is designed in detail, correction factors are derived and put back into the multi-period MINLP optimisation step.

5.2.7. Iterative Algorithm

The area of each of the heat exchangers obtained from the detailed network design is then compared to the areas that were predicted by the shortcut models from the multi-period MINLP step. The differences are used to calculate the correction parameters. $CorP_{i,j,k}$ is calculated by dividing the area of the detailed individual exchanger design by the area of the corresponding exchanger from the multi-period MINLP solution network. $U_{i,j,k,p}$, which is period dependent, is given the new values from the detailed design. $NSP_{i,j,k}$, the number of shell passes for the match, derived from the detailed designs, is also now inputted into the multi-period MINLP model for the respective exchanger. $XNSP_{i,j,k,p}$ refers to the number of shells that will be required for any extra exchangers that may be needed in the multi-period MINLP network. These four correction parameters are, importantly, limited to a change between runs to avoid drastically altering the solution space and potentially excluding solutions. In the case of this study, that change was limited to 5% between each iteration. Note also that this approach will result in non-integer values for $NSP_{i,j,k}$ and $XNSP_{i,j,k,p}$ between iterations. These values will either converge upon integer values by the end of the algorithm, or the non-integer values will "guide" the MINLP optimisation into not selecting the matches in further iterations.

Individual treatment is required for the remaining two correction parameters, $xy_{i,j,k,p}$ and $XA_{i,j,k,p}$, since they are both initialised at a value of zero, as we make the assumption that no extra exchangers are required with the first multi-period MINLP model run. $xy_{i,j,k,p}$, the binary parameter that indicates the existence of extra exchangers, is initially at '0' and if an extra exchanger is selected its value should be '1'. The drastic change from '0' to '1' can easily result in the match never being selected again in later iterations, so it is relaxed between iterations so that it can take values between '0' and '1'. This allows for the solution space to be altered in a small way between iterations and results in fewer potentially missed solutions. In this paper the maximum change between iterations is limited to '0.05'. $XA_{i,j,k,p}$, on the other hand, represents the area of the extra exchanger. Since this is also initialised at zero and is always multiplied by $xy_{i,j,k,p}$, this correction just takes on the value of the area of the extra exchanger that will be necessary. The fact that $xy_{i,j,k,p}$ is limited to small changes means that it limits the effect of large changes to $XA_{i,j,k,p}$ on the solution space. In this way it is allowed to slightly penalise the objective function initially and, if extra exchangers are

needed for this match in subsequent iterations, the penalty to the objective function is increased.

Initial values for the correction parameters should be considered carefully. Their initial values should always be estimates that will underestimate the objective function so that potential networks are not excluded (i.e. high overall heat transfer coefficients and no extra exchangers). Once these correction parameters are obtained they are inputted as parameters into the multi-period MINLP model and it is solved again. If the network is the same as the previous solution and none of the correction parameters change then the process is terminated. If the network is changed, then this new network is re-designed using the procedure described in Section 5.2.2 and new correction parameters are obtained. This iterative procedure is shown graphically in Figure 5.2. Since the networks selected can involve stream splitting and different network topologies are selected during many of the iterations, it is not guaranteed that the correction parameters will converge. In fact, for large examples it is unlikely to converge, even after many iterations. While this can be seen as a disadvantage, it allows for many different potential networks to be evaluated and designed in fair detail. In addition, subsequent networks are evaluated on information that is taken directly from the previous iteration. Because of this, a maximum number of iterations should be specified, and the optimal solution should be chosen as the solution with the design having the lowest TAC out of all of the iterations. This TAC should be the TAC of the detailed network design and not of the multi-period MINLP solution. It is not possible to determine where the optimal solution will be found; and therefore the higher the maximum number of iterations the more likely it will be that an optimal solution will be found. While this solution cannot be shown to be a rigorously determined optimum, it can be shown to result in very good solutions that take into account design factors that have not been considered in previous studies for multi-period networks. The reason that good solutions are reached is because the correction parameters shift the solution space, providing many different initial points for the MINLP solvers, as well as guiding the multi-period MINLP models towards more realistic designs that take into account many factors related to an actual network design.

5.3 Case Study

The methodology described above is now demonstrated with a case study.

5.3.1 Case Study and Results

The methodology was applied to a case study taken from Verheyen and Zhang (2006) consisting of equal durations for three periods containing 3 hot streams, 4 cold streams, and 1 hot and 1 cold utility. The problem has also been solved by Jiang and Chang (2013), Isafiade et al. (2015) and Isafiade and Fraser (2010), and the problem data is shown in Table 5.2. A superstructure consisting of 4 stages in which process streams can exchange heat was used. Since other methodologies for multi-period operation have not taken into account stream properties, stream properties were assumed to be the same for all streams and is shown in Table 5.3. Note that since we have the specific heat capacity and density of each stream in Table 5.3, the volumetric flow-rates are known. Also note that, for the sake of simplification, the process stream to utility exchangers are not calculated in the detailed network synthesis, as also done by Ravagnani and Caballero (2007b) and Mizutani et al. (2003b) for single period networks, and that the overall heat transfer coefficients for these matches is assumed constant at 720 W/(m^2·K).

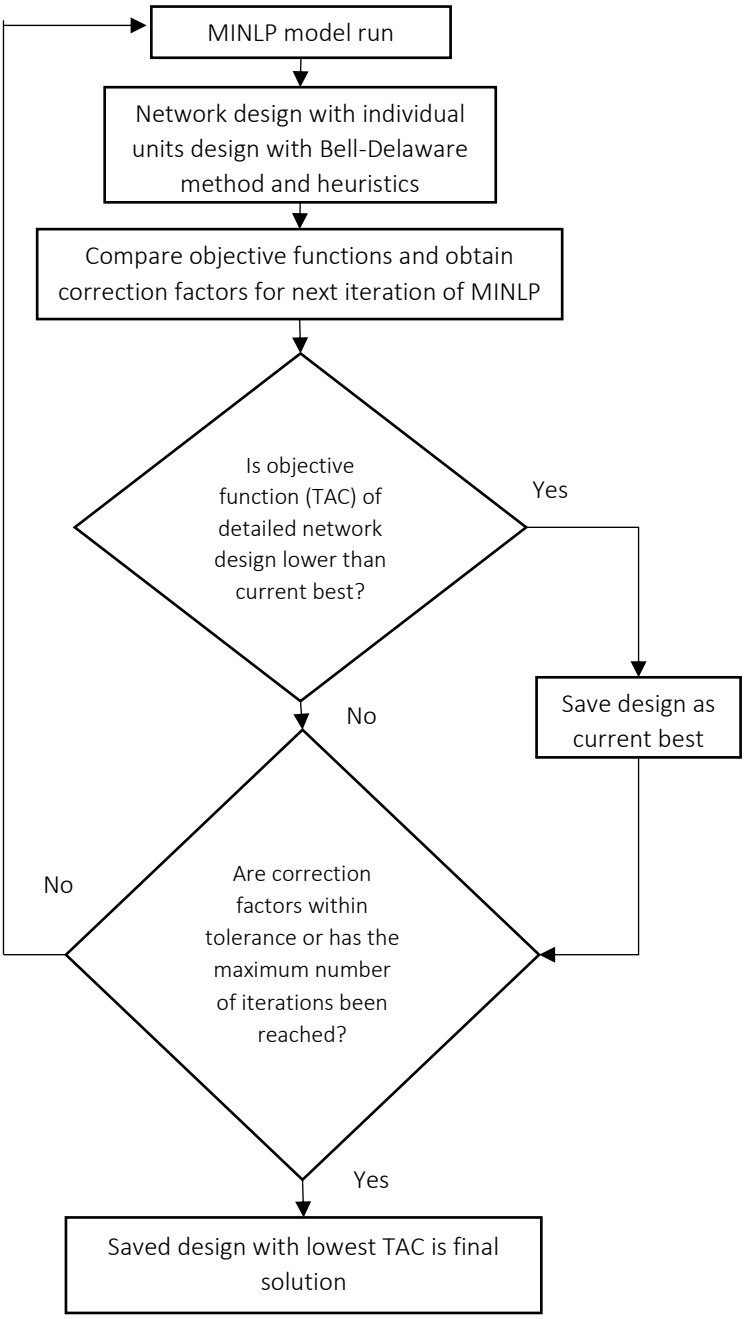

Figure 5.2: The iterative procedure used in this study.

Table 5.2: *Data for case study, taken from Verheyen and Zhang (2006)*

Streams	F (kW/ °C)	T_{in} (°C)	T_{out} (°C)
		Period 1	
Hot 1	201.6	393	60
Hot 2	185.1	160	40
Hot 3	137.4	354	60
Cold 1	209.4	72	356
Cold 2	141.6	62	210
Cold 3	176.4	220	370
Cold 4	294.4	253	284
		Period 2	
Hot 1	205.0	406	60
Hot 2	198.8	160	40
Hot 3	136.4	362	60
Cold 1	210.4	72	365
Cold 2	141.0	62	210
Cold 3	175.4	220	370
Cold 4	318.7	250	290
		Period 3	
Hot 1	208.5	420	60
Hot 2	175.2	160	40
Hot 3	134.1	360	60
Cold 1	211.1	72	373
Cold 2	140.5	62	210
Cold 3	174.5	220	370
Cold 4	271.2	249	286
		Utilities – all periods	
HU		500	450
CU		0	10

Where $\Delta_{min}T$ is 5 K, CF = 8333.3 \$/y, AC = 641.7, AE = 1, Cold utility cost = \$1.3/(kW.y), Hot utility cost = \$115.2/(kW.y), process-utilities overall heat-transfer coefficients = 720 W/(m^2.K)

Table 5.3: *Stream Data for all streams, taken from Mizutani, et al (2003b)*

μ (kg/(m·s))	ρ (kg/m^3)	c_p (J/(kg·K))	k (W/(m·K))	r_d (W/(m·K))
2.4E-4	634	2454	0.114	1.7E-4

All of the implicit correction parameters are given initial values for the first optimisation run that underestimate the objective function, as discussed in the methodology. The $NSP_{i,j,k}$ and $XNSP_{i,j,k,p}$ are initialised at 1 pass, $xy_{i,j,k,p}$ at 0, and $XA_{i,j,k,p}$ also at 0. $CorP_{i,j,k}$ is initialised at 1.05 because all exchanger areas are underestimated by at least 5% due to the LMTD approximation underestimating areas and the exclusion of F_T correction factors in the shortcut models. $U_{i,j,k,p}$ is initialised at 720 W/(m^2·K) as this is what the overall heat transfer coefficient for each match would be if the maximum velocities of the shell- and tube-side is used. Each MINLP optimisation consisted of 913 single equations, 941 single variables, and 140 discrete variables.

The method was carried out until 60 iterations was reached and the best solution was found after 57 iterations with a network consisting of 8 heat exchangers having 26 shells, 3 coolers, 3 heaters, 14 stream matches, and a final TAC of $3,020,475. This optimal network is shown in Figure 5.3, with the detailed exchangers and heat loads shown in Table 5.4 and network summary shown in Table 5.5. If we were to compare the solution to that that is obtained using Verheyen and Zhang's (2006) formulation (i.e. 1 run of the MINLP formulation) we would achieve an MINLP solution of $2,799,629, but when designing this network in detail, the true network cost is $3,068,946 (see Appendix 5B, Table 5B.2). In this network there is no need for any extra exchangers as a single exchanger is sufficient in each case across all of the periods. Exchanger 2 and Exchanger 4 have fairly high overdesign in some periods, however this is still well within the heuristic described in Section 5.2.5. Notice the extremely small exchanger, Exchanger 8. This exchanger was only selected in 1 period in the multi-period MINLP optimisation. It is designed here as a shell-and-tube exchanger, however, in practice this may be replaced with a double-pipe exchanger. Also note that in this example, only square tube arrangements were considered for the sake of simplicity, and with the assumption that this minimises pressure drops (and thus pumping requirements) as well as allows for easier shell-side cleaning.

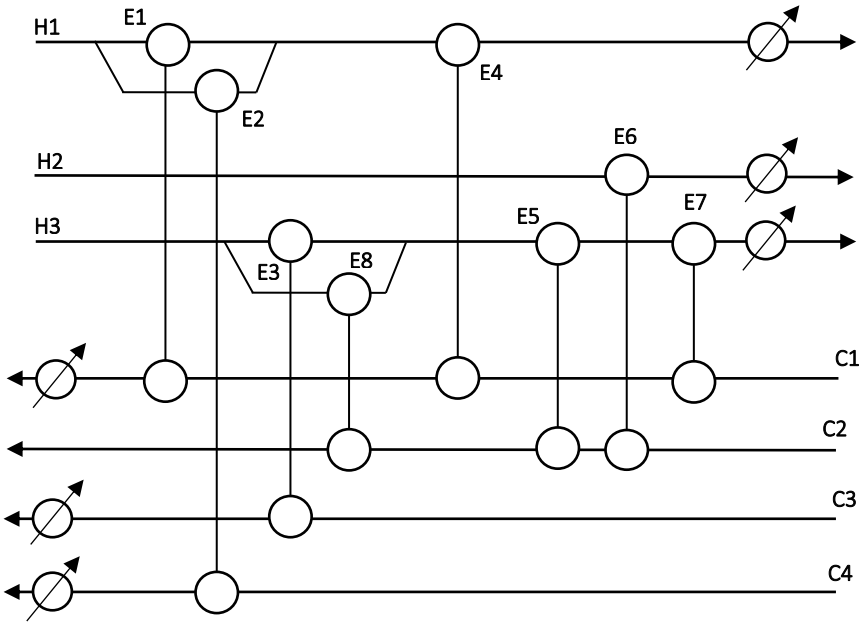

Figure 5.3: *Optimal network from case study*

Table 5.4: *Detailed designs for individual exchangers for optimal solution*

	E1	*E2*	*E3*	*E4*	*E5*	*E6*	*E7*	*E8*
Area (m^2)	1049.25	307.43	1213.67	3060.31	859.17	592.48	380.03	13.86
q (W)								
Period 1	19875.6	6856.87	16628.7	27759.1	10664.9	10291.9	7187.4	-
Period 2	20888.0	8593.1	17528.6	28270.8	10417.1	10450.9	7310.0	-
Period 3	21975.1	10034.3	16931.2	29613.0	10554.1	10028.1	6936.9	211.73
F_t								
Period 1	0.798	0.902	0.848	0.790	0.912	0.820	0.758	-
Period 2	0.791	0.925	0.845	0.796	0.919	0.832	0.76	-
Period 3	0.789	0.918	0.840	0.806	0.911	0.819	0.759	1.0
N_{sp}	3	2	5	7	5	2	1	1
N_{tp}	2	2	2	2	2	2	2	1
D_s (m)	0.889	0.889	0.7366	0.889	0.7366	0.889	0.8382	0.3366
N_t	1438	316	998	1438	942	812	1250	38
N_b	7	20	8	6	4	11	9	17
d_{ex} (mm)	15.875	31.75	15.875	15.875	15.875	19.1	15.875	31.75
d_{in} (mm)	11.66	27.53	11.66	11.66	11.66	14.83	11.66	27.53
pt (mm)	20.63	39.68	20.63	20.63	20.63	25.4	20.63	39.68
L (m)	4.876	4.876	4.876	6.069	3.658	6.069	6.069	3.658
$U_{overall}$ ($W/m^2 \cdot K$)								
Period 1	669.35	646.61	697.87	654.98	627.7	657.06	650.94	-
Period 2	651.37	687.31	699.93	654.23	624.6	659.61	649.69	-
Period 3	663.68	693.61	696.03	651.64	622.02	650.51	644.33	238.1
arr	Square	Square	Square	Square	Square	Square	Square	Square
Hot fluid allocation	Shell-side	Shell-side	Shell-side	Tube-side	Shell-side	Tube-side	Shell-side	Shell-side
TEMA Design	AES	AES	AEU	AES	AES	AES	AES	AES
Overdesign (%)								
Period 1	5.0	3.6	3.3	6.9	0.6	2.8	2.7	-
Period 2	1.23	12.6	2.9	8.3	5.2	4.3	3.2	-
Period 3	3.1	11.6	2.3	12.0	0.7	4.5	2.9	4.9

Table 5.5: *Solution summary for case study*

Solution summary	
Total annual cost ($/y)	3,020,475
Area cost:	
Annualised fixed area cost ($/y)	53,333
Annualised size area costs ($/y)	1,041,651
Utility cost ($/y)	1,925,490
Number of matches (including utilities)	14
Number of shells (including utilities)	32

It should be noted that a full list of the final values for all of the correction parameters is included in Appendix 5B in Table 5.B1. These values are not for a converged solution, but will allow the reader an insight into the kinds of values that the correction parameters lead towards. As can be seen there, a typical value for the $CorP_{i,j,k}$ is between 1.1 and 1.3. This means that in most cases, even with corrected $U_{i,j,k,p}$ values, the shortcut models used to

calculate the areas in normal MINLP network optimisation problems routinely underestimate the areas involved by between 10 and 30 %. With this knowledge, it should be noted that the $CorP_{i,j,k}$ were initialised at 1.05 (from the initial run) in order to speed up the convergence. In addition to this, the number of shells and the existence of extra exchangers is not a small matter to be ignored, with a very small number of exchangers actually utilising only a single shell. Interestingly, a solution that closely resembles the optimal solution was found at iteration 3 of the procedure. The only difference between this solution and the optimal solution found at iteration 57 is that the streams are split in slightly different ways and one of the matches required one extra shell.

The fact that the designer can guide the MINLP network optimisation by using corrections is useful in finding new designs and this allows for the designer to penalise the objective function for networks that require extra exchangers in order to carry out the heat load across all periods. In addition to this, the updated $U_{i,j,k,p}$ values that are dependent on the period of operation is important as well in determining whether the network can carry across the duty in all periods. At every iteration of the procedure, a network can be designed to any level of detail and evaluated by the designer for practical and financial aspects of the design. This will mean that the designer is not excluded from the optimisation procedure and can directly influence the optimisation by evaluating each network.

To demonstrate the usefulness of the approach, Figure 5.4 shows the difference between the objective functions of the multi-period MINLP optimisation and the detailed network design. As one can see, for the first 10 iterations, the multi-period MINLP optimisation underestimates the actual network costs by a large margin (more than 10 % in some cases). As the iterations increase there is a general trend that the difference in TAC between the actual generated network and the shortcut model solutions is lessened, with a higher chance of the optimisation model achieving a closer representation of a real design. Appendix 5B contains an abbreviated list of the networks generated with both objective functions displayed in Table 5B.2.

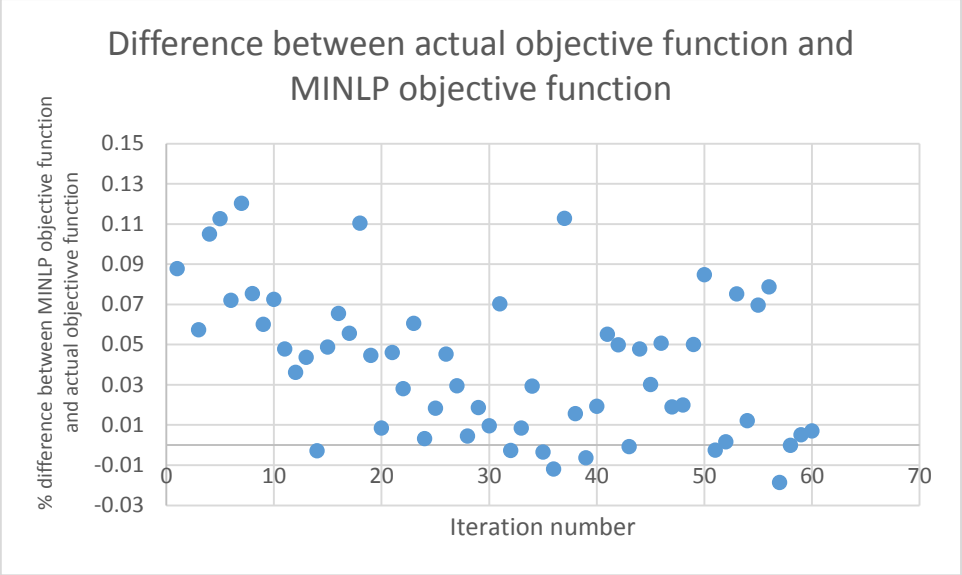

Figure 5.4: *Difference between the MINLP objective function and detailed network objective function*

In generating solutions for this problem, 60 iterations of the procedure was carried out and there was still no convergence. Although the use of a partial change of the correction parameters seems to increase the likelihood of reaching convergence. This is to be expected in a problem that involves a large number of binary variables as well as stream splitting. This is due to the fact that changes to correction parameters (even minor ones) can result in totally different networks and stream splits. Since the correction parameters are only valid for a specific stream split, they are then re-calculated and cannot converge upon a value as different networks are selected. Since the same match can be selected in different networks, it is unlikely that the correction parameters will converge, especially when considering stream splitting. This would be the case in most applications of using this kind of approach in mixed integer non-linear system. In order to demonstrate how extra exchangers may be needed, please refer to Appendix 5C, where an illustrative example is provided.

5.3.2. Remarks

The newly developed method is not limited to the model of Verheyen and Zhang (2006) for the optimisation step, and can use any simultaneous MINLP optimisation method, where all periods are simultaneously taken into account. The method is chosen here as it contains few non-linear constraints, increasing the chances of a globally optimum solution (though not guaranteeing it).

Similarly, it is not difficult to extend this model to include other aspects of multi-period design that may be of importance to practical designs, such as pressure drops, multiple exchanger types, network control and flexibility issues, as well as potential time-sharing mechanisms and cleaning schedules. While the case study in this paper is relatively simple with regards to stream composition, it is easy to include streams of varying composition. In addition, the fact that the design step and optimisation step are largely decoupled allows for the use of rigorous optimisation models like that of Ravagnani and Caballero (2007a) or commercial software such as AspenTech's EDR ™ or HTRI ™. The authors selected heuristic design with Bell-Delaware calculations so that the designer is able to decide whether a heat exchanger is suitable across multiple periods and to make a judgement on whether extra exchangers are necessary or whether excessive fouling may occur as a result of a flow-rate drop in one period or whether the pressure drop of a selected exchanger is too great to be considered.

One of the key elements in this methodology's success is the generation of many potential networks that can all be evaluated. The fact that the networks are generated in a "guided" way helps to find alternative structures that are likely to improve on some aspect of the design. The large number of initial points, provided by the correction parameters in each iteration allows for the generation of multiple starts that is more likely to find a global solution than other methods that attempt to bound and initialise the MINLP in intelligent ways.

One of the limitations of the work is that pressure drop has not been explicitly included in the optimisation, but only as a constraint when designing the individual heat exchangers. The inclusion of the highly non-linear pressure drop equations in the MINLP would result in numerical difficulties and would make finding feasible solutions more difficult. Future work is planned to overcome this issue. A further limitation is that the method is time-consuming due to the evaluation of the individual exchangers and their performance across all periods at each iteration. By utilising commercial software to evaluate the networks generated, it may be possible to overcome this limitation to a certain degree. Finally, the method cannot be guaranteed to converge on a final solution and therefore an arbitrary number of iterations is required, with the chances of finding an optimal solution increased as the number of iterations is increased. There is no way to determine when the optimal solution will be reached and whether the method will converge.

5.4. Conclusions

As has been noted throughout the study, the traditional approaches of synthesising multi-period heat exchanger networks encounter a number of issues when applied to real-life situations. The novel methodology presented in this work gets around this problem by using a detailed exchanger design with heuristics to "guide" the multi-period MINLP topology optimisation through the use of correction parameter that take into account details of the design that the shortcut models in the MINLP optimisation step are unable to account for, such as changes in overall heat transfer, TEMA (1988) designs, F_T correction factors, number of shell passes and tube passes, overdesigns, and the use of extra exchangers in certain periods if necessary.

The method's application to a case study shows its ability to generate a large number of potential solutions from which the best design can be chosen with regards to an objective function of a network designed in detail. The method is unlikely to converge upon a solution due to the presence of binary variables and stream splitting, which can result in very different networks being chosen between designs/iterations, however the method is successful in generating a large number of solutions that are "guided" by the presence of correction parameters, allowing for successively more realistic designs. The method cannot guarantee global optimality; however due to the many iterations with different initial points as a result of the correction parameters, it is more likely to find the global optimal solution than other methods. The inclusion of implicit correction parameters does not affect the non-linearity of the model but improves the accuracy of the shortcut models and allows for factors which account for design features such as extra exchangers and number of shells to be included into the objective function. The outer loop that includes heuristic design allows the designer to use experience and engineering judgement to find suitable designs that can be problem specific. The inclusion of a designer in multi-period network synthesis is particularly useful as aspects such as size restrictions, physical distances for piping, and control and flexibility can be assessed. The methodology can also be extended to include detailed exchanger optimisation and/or specialised software, such as AspenTech's EDR ™ or HTRI ™, as well as the inclusion of different exchanger types.

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5A. Appendix 5A: Exchanger Design Equations

Appendix 5A aims to clarify the methodology used to obtain the detailed designs for the methodology described in Section 5.3, above. Once the MINLP optimisation model is run, the stream properties and temperatures and energy balances obtained from the MINLP are inputted into an Excel spreadsheet. The equations and heuristics listed below are then used in order to design the largest exchanger required across all periods.

1. LMTD is calculated with:

$$LMTD = \frac{\Delta T_2 - \Delta T_1}{ln\left(\frac{\Delta T_2}{\Delta T_1}\right)}$$

Where ΔT_1 and ΔT_2 are the differences between the inlet temperature of the hot and outlet of the cold, and the inlet of the cold and outlet of the hot respectively.

2. F_T correction factors are calculated using the Blackwell and Haydu (1981):

$$R = \frac{T_{H,in} - T_{H,out}}{T_{C,out} - T_{C,in}}$$

$$S = \frac{T_{C,out} - T_{C,in}}{T_{H,in} - T_{C,in}}$$

$$F_T = \frac{\left[\frac{\sqrt{R^2+1}}{(R-1)}\right] \log\left(\frac{1-P_1}{1-RP_1}\right)}{\log\left[\frac{\left(\frac{2}{P_1}\right)-1-R+\sqrt{R^2+1}}{\left(\frac{2}{P_1}\right)-1-R-\sqrt{R^2+1}}\right]}$$

Where

$$P_1 = \frac{1-\left[\frac{R \cdot S-1}{(S-1)}\right]^{1/NS}}{R-\left[\frac{R \cdot S-1}{S-1}\right]^{1/NS}}$$

Where *NS* is the number of shells

Or, if *R* = 1,

$$F_T = \frac{P_2 - \sqrt{2}/(1-P_1)}{\log\left[\frac{\left(\frac{2}{P_1}\right)+\sqrt{2}}{\left(\frac{2}{P_1}\right)-\sqrt{2}}\right]}$$

Where

$$P_2 = P_1/(NS - NS \cdot S + P_1)$$

The *NS* is increased until the F_T is more than 0.75 (Kern, 1950).

3. U_0 (overall heat transfer coefficient) is estimated by the user and used to calculate area per shell:

$$A_o\ per\ shell = q/(NS \cdot U_o(F_T)(LMTD))$$

4. Tube size, shell size, thickness, length, pitch and configuration as well as head type are chosen using design heuristics from standard sizes according to heuristics detailed in Serth (2007).

5. These inputs are then used to get an estimate for the number of tubes, $n_{t,est}$, using:

$$(A_o)_{per\ shell}/(\pi d_{ex} L_t) = n_{t,est}$$

6. An estimate for velocity through the tubes is then used:

$$a_i = \frac{\pi d_{in}^2}{4}\left(\frac{n_t}{N_t}\right)$$

$$v_i = \frac{\dot{V}_i}{a_i}$$

N_t, the number of tube passes (either 1, 2, 4, 6, or 8), is increased in order to increase the velocity in the tubes to a value that is low enough to prevent excessive fouling and high enough to get good convective heat transfer (usually between 1 and 2 $m \cdot s^{-1}$ if the fluid is not water, with 1.7 $m \cdot s^{-1}$ being close to optimal) (Serth, 2007).

7. This estimate is then used to get the actual number of tubes, selected from an appropriate TEMA (1988) standards table (Serth, 2007). Once this is chosen, it is substituted into the equations above in the Excel spreadsheet to give an actual area to the exchanger. The velocity is then checked to see whether it is an appropriate choice. The shell diameter (D_S) is known from the TEMA standards.

8. A baffle cut is now chosen. 25% is used for all exchanger in the case for simplicity. The number of baffles, n_b, is chosen so baffle spacing (L_b), spacing ratio and the shell-side velocities (v_0) are all within the following:

n_b should be such that (L_B is baffle spacing $L_B = \frac{L_t}{n_B+1}$)

L_B> 50mm and

$L_B > 0.2\ D_s$

L_B < ½ X(max unsupported tube length)

Shell side velocity (v_o) is, for

Liquids 0.3-1.0 m/s

$v_o = \frac{\dot{V}_o}{a_o}$

where $a_o = \frac{(P_t - d_o)(D_s)L_B}{P_t}$

9. The inside heat transfer coefficient (h_i) is now calculated for the chosen exchanger using the modified Dittus-Boelter equation:

$$Nu_i = \frac{h_i d_{in}}{k_i} = 0.023 Re_i^{0.8} Pr_i^n \left(\frac{\mu_i}{(\mu_w)_i}\right)^{0.14}$$

Where n=0.3 for fluid being cooled and n=0.4 for fluid being heated. μ_i is considered equal to $(\mu_w)_i$ in the examples considered where $(\mu_w)_i$ is the fluid viscosity at the wall.

$$Re = \frac{\rho v D}{\mu}$$

$$Pr = \frac{\mu C_p}{k}$$

$$Nu = \frac{hD}{k}$$

10. Using d_o & P_t, ascertain the equivalent diameter (d_e) for the shell-side

$$d_e = \frac{4\left[S_L S_T - \frac{\pi d_{ex}^2}{4}\right]}{\pi d_o}$$

Where S_L=S_T=P_t for square pitch (used for the case study) and S_L= 0.866P_t, S_T=P_T for equilateral triangular pitch.

And then derive the outside heat transfer coefficient using

$$Nu_o = h_o \frac{de}{k_o} = J_h Re_o (Pr_o)^{\frac{1}{3}} \left(\frac{\mu_o}{(\mu_w)_o}\right)^{0.14}$$

J_h is the heat transfer correction factor from the Bell-Delaware method and is determinedgraphically as prescribed by the Bell-Delaware method (Serth, 2007)

11. Using the h_o and h_i, fouling factors and tube diameters, the overall heat transfer coefficient is found using:

$$\frac{1}{(U_o)_{actual}} = \left(\frac{d_o}{d_i}\right)\left(\frac{1}{h_i}\right) + \frac{d_o \ln\left(\frac{d_o}{d_i}\right)}{2k} + \frac{1}{h_o} + \frac{d_o}{d_i}R_i + R_o$$

12. The actual required area of the exchanger can now be found:

$$(A_o)_{required} = \frac{q}{(U_o)_{actual}(F_T)(LMTD)}$$

The design is then assessed as to whether it is over- or under-designed.

13. Once all of this information is inputted into a spreadsheet, it is possible to quickly and efficiently create good designs. When the single largest exchanger across all periods is designed then the requirements across the other periods is tested. The heuristics mentioned above in Section 5.2.5 are all checked. If the design is suitable across all periods then the exchanger is chosen. If there is a period in which the design is not suitable for any reason then another exchanger is designed for this period. The pressure drop can also be determined at this stage using methods outlined in Serth (2007) and should be checked as to whether it is above a certain threshold (50 kPa per shell was chosen for this study). The extra exchanger will be smaller than the main exchanger and will penalise the objective function, so it is important for the designer to attempt to avoid this situation. A good designer is able to use these heuristics with common sense and knowledge of the process fluids to obtain near-optimal designs for a given circumstance.

5B. Appendix 5B: Supporting Information

This appendix contains supporting information for the case study.

Table 5B.1 should be read as follows. Every “match” corresponds to (hot stream, cold stream, interval). The process stream to process stream intervals begin at temperature location ‘2’ and end at temperature location ‘5’. Whenever a value is absent it is at its initial value (as detailed in Sections 5.2.3 and 5.3). When a value is present and at its initial value, this means that it was changed from its initial value at some point during the algorithm, however by iteration 60 (the final iteration) it has returned to its initial value.

Table 5B.1: *Values for all correction factors at final iteration*

Match	$CorP_{i,j,k}$	$U_{i,j,k,p}$	$NSP_{i,j,k}$	$xy_{i,j,k,p}$	$XA_{i,j,k,p}$	$XNSP_{i,j,k,p}$
('1','1','2')	1.25223	0.6872 0.660705 0.66243	2.3767	0.0 0.0	128.4 912.07	1.1025 1.05
('1','3','2')	1.248	0.60386 0.601 0.613	1.7735	0.15 0.0	317.4 203.14	1.05 1.2025
('1','4','2')	1.0921	0.672 0.703 0.68085	1.0807	0.6 0.0 0.0	13.86 31.01 134.26	1.0 1.0 1.0
('3','1','2')	1.267	0.66545 0.66316 0.64669	1.33	0.0	1116.9	1.1575
('3','2','2')	1.1025	0.684				
('3','3','2')	1.2625	0.68766 0.68425 0.67682	1.98	0.0	1431.96	1.1576
('3','4','2')	1.153	0.65717 0.690144 0.67373	1.27	0.25	350.24 363.1;	1.0 1.0
('1','1','3')	1.31175	0.681 0.683 0.685	4.9153	0.0	252.46 238.4 620.21	1.336 1.1575
('1','2','3')	1.05	0.6878 0.6866 0.684	1.05			
('1','3','3')	1.2696	0.676 0.68393 0.6972	1.536	0.0	42 109	1.0 1.0
('1','4','3')	1.1515	0.6302 0.620832 0.679	1.2064	0.15	150.02 105.44 116.75	1.0 1.0 1.0
('3','1','3')	1.23288	0.65207 0.6235 0.63998	1.5475	0.05	 572.78	 1.05
('3','2','3')	1.257	0.638214 0.63547 0.62867	1.98			
('3','3','3')	1.265	0.66207 0.66036 0.65466	2.1	0.0	56.2 728.2 558.43	 1.1575

('3','4','3')	1.2936	0.68469 0.651 0.6754	1.05	0.05	363.08	
('1','1','4')	1.38	0.608 0.611 0.622	3.033	0.0	168.5 240.8	1.05 1.05
('1','2','4')	1.2245	0.62423 0.62303 0.61508	1.7913	0.0	916.45 255.4	1.1025
('1','3','4')	1.2346	0.65515 0.65992 0.6632	1.273	0.0	112.8	
('1','4','4')	1.1576	0.65 0.67807 0.6755	1.1025			
('2','1','4')	1.116	0.67008 0.69247 0.6586	1.4057	0.0	401.9 624.6	1.05
('2','2','4')	1.24	0.65706 0.6596 0.65051	1.4036	0.0	 273.2	 1.05
('3','1','4')	1.31314	0.67262 0.66999 0.6638	2.1661			
('3','2','4')	1.346	0.648 0.646 0.645	1.886			
('3','3','4')	1.19199	0.687663 0.6895 0.6854765	1.5512	0.0	 533.14	 1.05
('1','1','5')	1.2545	0.67773 0.67291 0.685782	2.298			
('1','2','5')	1.1641	0.675758 0.67335 0.67453	1.393	0.0	 175.9	 1.0
('2','1','5')	1.1605	0.6566 0.67827 0.6408875	2	0.0	 189.75	 1.0
('2','2','5')	1.1816	0.637 0.638 0.631	1.6288	0.0	340.5	1.05
('3','1','5')	1.3953	0.61396 0.624815	1.4774			

		0.610708				
('3','2','5')	1.4011	0.6386 0.6355 0.63673	2.2863	0.05	1820.5	1.1025

Table 5B.2: *Summary of networks evaluated during the case study*

Iteration	MINLP objective function ($/year) (a)	Detailed design objective function ($/year) (b)	Ratio (a/b)	Number of matches	Number of Shells
1	2799629.622	3068946.481	0.912245	13	34
2	2926896.201	3493838.334	0.837731	12	38
3	2868363.512	3042928.093	0.942633	14	33
4	2852602.039	3187202.108	0.895018	15	36
5	2907616.757	3276687.106	0.887365	13	42
10	3058455.67	3297779.958	0.927429	14	44
20	3018499.174	3044168.286	0.991568	14	34
30	3089741.591	3119506.867	0.990458	11	31
36	3069445.738	3033528.203	1.01184	15	39
40	3274723.359	3339044.564	0.980737	12	46
50	3161422.715	3454150.951	0.915253	13	46
51	3165071.923	3157389.087	1.002433	10	31
57	3076817.42	3020475.131	1.018653	14	32
58	3067270.427	3067125.3	1.000047	14	33
59	3162184.914	3178393.361	0.9949	13	35
60	3201116.093	3223924.712	0.992925	10	30

5C: Appendix 5C: Illustrative Example

This appendix contains an illustrative example of an extra exchanger being required. Since the optimal solution for the example considered in this paper contained a network that did not require an extra exchanger, this section aims to illustrate how the extra exchangers are determined and in what case they would be necessary. In iteration 2 of the solution process a network is generated that contains the match (‘4’, ‘3’, ‘3’) which is an exchanger that is required to function in all 3 periods. The detailed exchanger design for this match is shown in Table 5C.1.

***Table 5C.1**: Illustrative case for an extra exchanger*

	Period 1	*Period 2*	*Period 3 (in representative exchanger)*	*Period 3 (in extra exchanger)*
Area (m^2)	837.65	837.65	837.65	687.34
q (W)	7525.58	7291.55	6940.03	6940.03
F_t	0.851	0.849	0.880	0.880
N_{sp}	3	3	3	3
N_{tp}	2	2	2	2
D_s (m)	0.787	0.787	0.787	0.7366
N_t	1148	1148	1148	942
N_b	6	6	6	6
d_{ex} (mm)	15.9	15.9	15.9	15.9
d_{in} (mm)	11.66	11.66	11.66	11.66
pt (mm)	20.6	20.6	20.6	20.6
L (m)	4.877	4.877	4.877	4.877
$U_{overall}$ ($W/m^2\,^oC$)	609.14	602.82	591.12	617.7
arr	Square	Square	Square	Square
Hot fluid allocation	Shell-side	Shell-side	Shell-side	Shell-side
TEMA Design	AES	AES	AES	AES
Overdesign (%)	5.0	3.6	17.9	1.1

In Table 5C.1 it is evident that if the same shell configuration is utilised over all periods it will be ineffective for period 3, with an overdesign of 17.9%. Attempting to use a smaller exchanger will result in an under-designed exchanger for periods 1 and 2, meanwhile an overdesign of that degree for period 3 will result in problems further down- or upstream as well as in the operation of the unit. In order to accommodate the design proposed by the MINLP optimisation, an extra exchanger is thus designed that can be used in period 3 without any problems. As a result of this extra exchanger, the overall cost of the network is increased due to the extra capital expenditure, meanwhile operability and flexibility is increased. In order to account for this extra exchanger in subsequent iterations, the corrections $xy_{i,j,k,p}$, $XA_{i,j,k,p}$, and $XNSP_{i,j,k,p}$ are incrementally increased in order to penalise the selection of that match in subsequent iterations. In some cases it is also possible that in one period the F_T correction factor is too low for a specific period and an extra exchanger, to provide the correct number of shells, is required in a single period.

Chapter 6

Mass Exchanger Network Synthesis

Chapter 6: Mass Exchanger Network Synthesis

This chapter is a verbatim reproduction of a paper that will be submitted for review to Chemical Engineering Science in June 2017. The paper is entitled “Synthesis of Mass Exchanger Networks in a Two-Step Hybrid Optimisation Strategy Involving MINLP Network Synthesis and Detailed Packed Column Design”. As with Chapters 4 and 5, the changes made are only cosmetic so as to keep the formatting and numbering consistent throughout the thesis. Also, as with the other chapters, all of the work and writing within the paper are the original contributions of the current author, with all other authors’ supervision acknowledged. While the previous chapters have dealt with the implementation of the proposed strategy to HENS, this chapter deals with the implementation to MENS. The MENS MINLP formulation is more difficult to solve than the HENS formulation, due partly to the small numbers involved when calculating the compositions, and also due to the additional costing considerations involving diameter and volumes. This chapter is particularly interesting due to the novel NLP sub-optimisation developed within it that uses orthogonal collocation on finite elements to rigorously optimise the detailed designs of the columns, including the packing sizes. This differs from the previous two chapters that have involved a heuristic and manual approach to the detailed unit designs, allowing for a much faster generation of the designs, and also a rigorous search for the optimal detailed mass exchange network. The approach, along with the pertinent literature follows.

Synthesis of Mass Exchanger Networks in a Two-step Hybrid Optimisation Strategy Involving MINLP Network Synthesis and Detailed Packed Column Design

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Abstract

This study presents a new method for the synthesis of mass exchanger networks (MENs) involving packed columns. Simultaneous synthesis of MENs is typically done through the use of mixed-integer nonlinear program (MINLP) optimization, with simplifications made in the mathematical representations of the exchangers due to computational difficulty in solving large non-convex mixed-integer problems using current solver technology. These simplifications include use of fixed mass transfer coefficients, fixed column diameters, no considerations for pressure drops, and iso-compositional mixing for models that use the stage-wise superstructure is usually adopted. The methodology proposed in this study also makes use of the stage-wise superstructure MINLP formulation for the network synthesis. However, it includes a detailed individual packed column design in a non-linear programming (NLP) sub-optimisation step, where orthogonal collocation is utilized for the partial differential equations, and that finds the optimal packing size, column diameter, column height, fluid velocities, etc., in relation to a cost-based objective function. The detailed designs are then used to determine correction factors that update the simplified models to more accurately portray the chosen design. Once the MINLP is updated with these correction factors, the model is re-run, with new correction factors obtained. This iterative procedure is repeated until convergence between the objective function of the MINLP and that of the NLP sub-optimisation is achieved, or until a maximum number of iterations is reached. The methodology is applied to several examples and is shown to be robust and effective in generating new topologies, and in finding networks that are physically realizable.

Keywords: mass exchanger networks; MEN; non-linear programming; mathematical programming; orthogonal collocation; packed columns

Nomenclature

Abbreviations

ACC	Annual capital cost
AOC	Annual operating cost
HENS	Heat exchanger network synthesis
IBMS	Interval based MINLP superstructure
LMCD	Logarithmic mean composition difference
MENS	Mass exchanger network synthesis
MSA	Mass separating agent
MINLP	Mixed integer nonlinear program
NLP	Nonlinear program
OC	Orthogonal collocation
OCFE	Orthogonal collocation on finite elements
SBS	Supply-based superstructure
SWS	Stage-wise superstructure
TAC	Total annual cost

Sets

R	Rich process streams
S	Lean process and external streams
INT	Superstructure intervals

Indices

r	Process rich streams
l	Process lean and external lean streams

k Index representing interval, $1, \ldots \ldots . NOI$ and composition location, 1,.....$NOI + 1$

ii Collocation point

jj Finite element

tt Total number of gridpoints over the entire height of the column

Parameters and variables

$ai_{r,l,k}$ Packing specific surface area $(m^{-2}m^{-3})$

$aiCor_{r,l,k}$ Packing specific surface area correction factor

AC_l Annual operating cost per unit of lean stream l ($/(kg.y))

AF Annualisation factor

$FC_{r,l,k}$ Installation/fixed cost for mass exchanger r, l, k ($)

$Cl_{r,l,k,ii,jj}$ Concentration of component in the lean stream in match r, l, k at collocation point ii and finite element jj (*kg/m^3*)

$Cr_{r,l,k,ii,jj}$ Concentration of component in the rich stream in match r, l, k at collocation point ii and finite element jj (*kg/m^3*)

$D_{r,l,k}$ Diameter of column r, l, k (m)

$Dcor_{r,l,k}$ Diameter correction factor for match r, l, k

Vis_r Viscosity of of liquid stream l $(Pa\ S)$

$Flux_{r,l,k,ii,jj}$ Flux of component from rich stream to the lean stream in match r, l, k at collocation point ii and finite element jj (gs^{-1})

$FloodPoint_{r,l,k}$ Pressure drop at which flooding is likely to occur

FP Column packing factor

G_r Flowrate of rich stream (gs^{-1})

$Hcor_{r,l,k}$ Height correction factor for match r, l, k (m)

He Henry's coefficient

$ky_{r,l,k}$ Overall mass transfer coefficient for match r, l, k, $(gs^{-1}m^{-3})$

$kyCor_{r,l,k}$	Overall mass transfer coefficient correction factor for match r, l, k
NC	Number of collocation points
NE	Number of finite elements
$PackCost_{r,l,k}$	The cost of packing ($\$/m^3$)
$PackDens_{r,l,k}$	The density of the packing (kgm^{-3})
$Pdrop_{r,l,k}$	Actual pressure drop (kPa/m)
$RHOL_{r,l,k}$	Lean stream density for match r, l, k (gm^{-3})
$RHOG_{r,l,k}$	Rich stream density for match r, l, k (gm^{-3})
$SA_{r,l,k}$	Surface area of packing (m^2/m^3)
u	Normalized spatial variable (defined in Equations 8 and 9)
X_l^s	Supply composition of lean (process or external) stream
X_l^t	Target composition of lean (process or external) stream
Y_r^s	Supply composition of rich process stream
Y_r^t	Target composition of rich process stream
Y_l^{*s}	Equilibrium supply composition of lean (process or external) stream
Y_l^{*t}	Equilibrium target composition of lean (process or external) stream
σ_c	Critical surface tension
σ_l	Surface tension of the liquid stream
Ω	Exchanged mass upper limit for match r, l, k
Φ	Driving force upper limit for match r, l, k
π	Mathematical constant = 3.142
$\mu_{r,l,k}$	Viscosity of the gas stream in match r, l, k $(Pa.s)$
Sc	Gas Schmidt number
ω	Function of the wetted packed surface
a_G	Experimental constant which is a function of packing
β	Experimental constant which is a function of the packing

d_e Equivalent packing diameter (m)

$\in$ Fractional voidage of the column packing

$G'_{r,l,k}$ Superficial gas mass velocity in match r, l, k $(gm^{-2}s^{-1})$

Binary Variable

$Z_{r,l,k}$ Represents the existence of match r, l in interval k in the optimal network

Positive Variables

$A_{r,l,k}$ Cross-sectional area of column r, l, k (m)

$FLV_{r,l,k}$ Flow factor for column r, l, k

$KF_{r,l,k}$ Flooding parameter for column r, l, k

$Fr_{r,l,k}$ Rich stream split branch flowrate for column r, l, k (kgs^{-1})

$Fl_{r,l,k}$ Lean stream split branch flowrate for column r, l, k (kgs^{-1})

$H_{r,l,k}$ Packed height for column r, l, k (m)

$dy_{r,l,k}$ Mass exchanger driving force

L_p Liquid flowrate at periphery of packing, $m^3s^{-1}m^{-1}$

L_l Flowrate of lean stream (kgs^{-1})

$LMCD_{r,l,k}$ Logarithmic mean composition difference between rich stream *r* and lean stream l in interval k

$M_{r,l,k}$ Mass exchanged between rich stream *r* and lean stream l in interval k (kgs^{-1})

$y_{r,k}$ Composition of rich process stream in composition interval boundary k

$x_{l,k}$ Composition of lean (process or external) stream in composition interval boundary k

$y^*_{l,k}$ Equilibrium composition of lean (process or external) stream l in composition interval boundary k

6.1 Introduction

Focus on decreasing the chemical industry's impact on the natural world and human health has been a key feature of the modern era of industrial development. In order to meet new emissions standards, decrease the threat of climate change, and reduce the impact of the chemical industry on human health and ecological systems, pollutant reduction is a necessity. In many cases, streams are required to be treated in order to discharge waste with acceptable levels of dangerous chemicals. Mass exchanger networks (MENs) are often employed to fulfil this task, using either other process streams or mass separating agents (MSAs) to absorb pollutants from streams to be discharged. Traditionally MENs have been designed with pinch technology (PT), but these methods have the disadvantage that they are sequential in nature, with design targets being set and heuristics employed in order to find networks that approach or meet the targets. With the proliferation of computers and increased efficacy of non-linear and mixed-integer solvers, simultaneous approaches have gained momentum, having been shown to be more adept at finding better solutions, and superior counter-intuitive solutions. Unfortunately, solver technology is still limited in its ability to guarantee globally optimal solutions for non-convex and mixed integer problems, meaning that large problems are difficult to solve and are often reformulated using simplified shortcut models to represent the individual exchangers. In so doing, the resulting network is often not physically attainable or the individual units are over- or under-designed. Examples of typical simplifications that have been employed include fixing diameters for all columns involved, simplifying capital costing equations, fixing the column internals' dimensions, ignoring pressure drops and flooding considerations, and fixing mass transfer coefficients for specific stream pairs.

6.2 Literature Review

6.2.1 Mass Exchanger Network Synthesis (MENS)

The MENS problem was first defined in a paper by El-Halwagi and Manousiouthakis (1989). In this work they extended the pinch approach for heat exchanger network synthesis (HENS) to the application of mass exchange networks by first targeting the minimum cost of MSAs without a predetermined network structure and then, in a second stage, minimizing the fixed cost by generating a network with a minimum number of exchangers. Hallale and Fraser (2000a) extended the methods of El-Halwagi and Manousiouthakis (1989, 1990a, 1990b) to include capital costs targets. Through the inclusion of capital cost targets assumptions that the optimal capital costs could be found by targeting the minimum number of exchangers or trays above and below the pinch was avoided and a more realistic and useful optimisation could be undertaken. This also enabled the use of super-targeting for the first time in MENS, where the optimal minimum driving force could be found by trading off both capital and operational costs while varying the minimum driving force. Hallale and Fraser (2000b) extended upon this in a subsequent paper to include more detailed capital costing for both packed and staged columns (Hallale & Fraser, 2000a, 2000b, 2000c).

These approaches, while effective, are sequential in nature and therefore fail to take into account all the variables involved in the design of a network. A simultaneous mathematical programming approach was first used by Papalexandri, et al. (1994). Their study formulated the MENS problem as a mixed-integer nonlinear programme (MINLP) where the network hyperstructure approach, developed by Ciric and Floudas (1989) for HEN synthesis, was adapted for MENS. El-Halwagi (1997) notes that this approach excludes solutions by excluding certain configurations, excludes the designer from the design, and can also potentially find only locally optimal solutions due to non-convex terms. Comeaux (2000) simplified the model formulation presented by Papalexandri, et al. (1994), adopting pinch principles to formulate the MENS problem as an NLP of moderate size.

Most of the modern attempts at finding truly optimal solutions have been adapted from the stagewise superstructure (SWS) approach of HENS (Yee & Grossmann, 1990). In this approach a superstructure is constructed that attempts to embed the most possible potential network topologies within it. Szitkai, et al. (2006) used a similar approach to Yee and Grossmann's (1990) approach to HENS in the synthesis of MENs. Their model was

formulated as an MINLP and they presented a mostly linear single contaminant model as well as proposing a multiple contaminant model with the addition of nonlinear constraints. The authors commented that their approach was best suited to single contaminant problems with packed columns as mass exchangers (Szitkai, et al., 2006). They made use of the integer-infeasible path MINLP (IIP-MINLP) (Sorsak & Kravanja, 2002) model formulation to add stability and enhance the search for numerical solutions. Their model made the assumption that mixing streams must have the same compositions, limiting the solution space, but removing non-linearities. Emhamed, et al. (2007) introduced a hybrid approach to MENS that combined both Hallale and Fraser's (2000a) supertargeting approach with the MINLP optimization strategy of Szitkai, et al. (2006). The method uses integer cuts and a bounding strategy in order to improve on the solutions generated in subsequent iterations and avoid infeasible networks. Isafiade and Fraser (2008) presented a novel method of setting up superstructures in MENS, named the interval-based MINLP superstructure (IBMS) model. The model defines the stages of the superstructure in terms of the supply and target compositions of the rich and lean streams. The authors found that the new method provides different solutions to the model of Szitkai, et al. (2006) in some cases providing better solutions; showing that the solutions of Szitkai, et al. (2006) are most likely locally optimal underlining the problems associated with MINLP solvers. The authors suggested that, by defining superstructure stages in this way, it is possible that the problem is more efficiently initialized and bounded, resulting in more efficient solution times (Isafiade & Fraser, 2008).

Azeez, et al. (2012) and Azeez, et al. (2013) extended on the IBMS by exploring other possible interval-based options. Their study was extended to both HENS and MENS and defined the intervals by supply and target compositions in different ways, eventually resulting in the supply and target based superstructure (S&TBS), target and supply based superstructure (T&SBS), and the supply based superstructure (SBS). These new superstructure formulations were applied to a host of examples and it was found that by re-formulating the superstructure in this way, new solutions could be found, however there was no conclusion as to which of the approaches provides consistently good solutions. The methods were applied to both staged and packed columns (Azeez, et al. (2012), and Azeez, et al. (2013)).

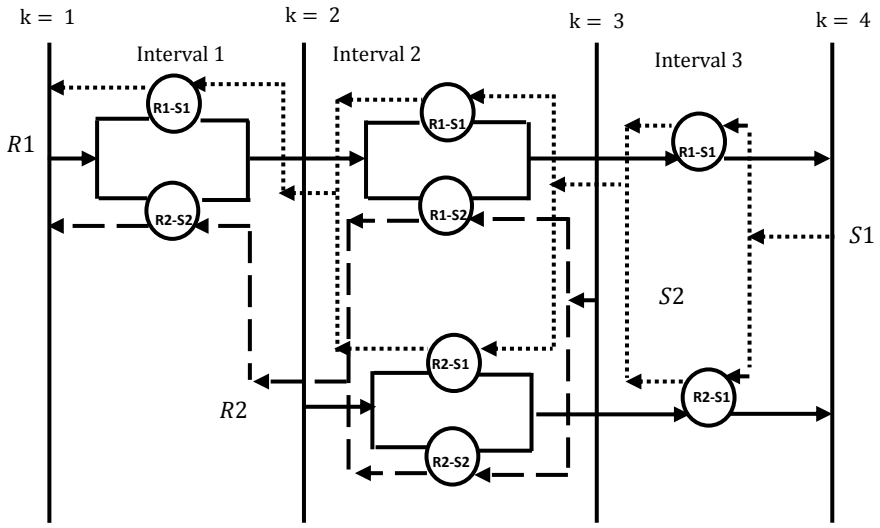

Figure 6.1: *Supply based superstructure (SBS) used in this paper, adapted from Azeez, et al. (2012)*

Liu, et al. (2013) used an NLP formulation to find optimal MENs with multiple components and used a genetic algorithm-simulated annealing algorithm hybrid approach (GA-SA). Their method does not rely on key components and uses number of trays to account for capital costing. They present only a small example that fails to find the best solution, even with a simplified superstructure in their formulation (Liu, et al., 2013).

In all of these mathematical programming based methods for designing networks of mass exchangers, the columns are approximated with equations that do not necessarily represent the final design. The fixing of column diameters, fixing of mass transfer coefficients such that it does not depend on fluid velocities, disregard for flooding conditions in terms of possibility of flooding or otherwise, and inability to accurately select from different packing parameters mean that there is little scope for the methods to be applied to industrial examples. This is possibly why sequential optimization methods are still popular in industry today. The only attempt at including these details came in the way of Isafiade and Short (2016).

Isafiade and Short (2016) extended the SBS model of Azeez, et al. (2012) by including the diameter as a variable in the MINLP. With this, it was also possible to include the overall mass transfer coefficient as a variable for each stream by the inclusion of Pratt's correlation (Leva, 1953) by considering velocities of both rich and lean streams. With these inclusions it was thus necessary to also consider the column's packing and flooding potential. Due to the highly non-linear nature of the additional equations, a special initialization strategy was proposed to get initial solutions. Once the solution was generated the resulting columns

were checked whether flooding was likely to occur or not. If a column was detected to flood, a new packing was selected for a subsequent run and the model re-run. The model was successfully applied to both single and multi-period examples, however the highly nonlinear nature of the additional equations and the subsequent manual flooding check and sequential changing of packing parameters only for the columns that were likely to flood meant that there was no guarantee of a globally optimal solution and the model proved difficult to initialize and find feasible solutions. A further disadvantage is that the resultant columns proved to be quite unrealistic with large diameters and small heights.

It should be noted that the model of Isafiade and Short (2016) was not used in the current study due to the added nonlinearities that resulted in difficulty in finding feasible solutions and the addition of manual initialization stages. While this model includes many more detailed aspects at the topology level, it is difficult to solve and therefore is likely to be trapped in local optima. In addition to this, there was no way to directly include flooding considerations, or the inclusion of particular packing sizes, and these had to be included in a subsequent manual step, with no way of ensuring that the chosen parameters were optimal.

While it is evident from the literature surveyed that there is a lack of focus from the process synthesis community on MENS, much research has been done on the HENS problem in the last 20 years. Many of the advancements in MENS have indirectly come from the HENS research, and the current work also takes inspiration from HENS. Short et al. (2016a) made use of a novel method for the synthesis of HENs whereby the initial network synthesis was achieved by an MINLP model similar to that of Yee and Grossmann's (1990) SWS. Noting that the equations in the SWS contain approximations of the heat exchanger areas, as well as the unrealistic fixing of heat transfer coefficients, the lack of pressure drop considerations, or penalties for the inclusion of multiple shells, the author's then used more accurate heat exchanger heuristic models to design the network in detail. They found that the rigorously designed exchangers differed drastically from the MINLP shortcut models' approximations. In an attempt to help the MINLP find solutions that were more realistic the authors included a host of correction factors that forced the MINLP shortcut models to converge upon the solution of the more rigorous design equations after a number of iterations between the MINLP model and rigorously designed versions of the MINLP model solutions. The resulting networks were thus physically attainable in a real design scenario and also were able to find new networks that used the detailed design information without including the highly nonlinear detailed design equations in the MINLP. In addition to this, the authors noted that

during the iterative process many new initial points were used as a result of the constant updating of correction factors, meaning that the MINLP optimization solution was more likely to find globally optimal solutions than other methods over the course of the algorithm. The authors also extended the approach to multi-period HENS, showing the versatility and robustness of the approach (Short, et al., 2016b). The authors of this current paper look to extend this work from the field of HENS to that of MENS. This would be accomplished through the inclusion of correction factors to the MINLP stage, followed by a more detailed NLP optimization of the individual packed columns, and using this information to update subsequent runs of the MINLP topology optimization. In this new formulation, presented below, the inclusion of detailed packed column optimization, as opposed to the heuristic, manual designs used in the study of Short, et al. (2016a), allows for more rigorously determined optimal designs for both the network and the individual columns, as well as the ability to automate the process in order to quickly determine the optimal detailed network for a given problem.

6.2.2 Individual Mass Exchanger Optimisation

While much work has been done on optimizing the design of trayed columns, packed column design has received less attention. The most common approach to designing gas-liquid packed columns has been through dividing the column into a number of equilibrium stages, with the concentration profiles determined by assuming the equilibrium state of the gas and liquid streams leaving each equivalent stage. The method, known as HETP (Height Equivalent to a Theoretical Plate), has no theoretical basis (Seader & Henley, 1998) and since the packed column is a continuously contacting unit, it is best treated as such. Due to the nature of random packings however, it has so far not been possible to avoid the use of empirical correlations entirely.

There have been numerous steady state and dynamic simulations of packed columns. Since the detailed models include partial differential equations, a number of different approaches have been used in attempting to solve these equations. The most common method of solving sets of partial differential equations in engineering is through the use of orthogonal collocation on finite elements (OCFE). It has been shown by various authors (Karacan, et al, 1998; Biegler & Logsdon, 1989) that the use of OCFE can drastically decreased the CPU time needed and shows sufficient accuracy in approximating differential equations.

Numerous studies have shown that it is possible to simulate complex reactive absorption packed columns to a relatively high degree of accuracy through the use of mass and energy balances through the use of either HETP or through solving sets of differential equations. Srivastava and Joseph (1984) used polynomial approximation techniques and applied 2-film theory in their solution of a packed column. Their model, validated through experimentation, found that accurate solutions could be found with fewer than 4 collocation points. The model had fixed parameters for the column, but solved for the height of packing. The study showed that collocation techniques greatly reduce the number of equations in comparison with finite difference techniques.

Typically, detailed distillation columns have been modelled using the technique of Krishnamurphy and Taylor (1985). In their technique, mass and energy balances are performed over a number of elements in terms of each phase, with a non-equilibrium stage model that assumes equilibrium conditions at the interface. This is then solved simultaneously using Newton's method. Their method forms the basis for Aspen's RADFRAC ™ model, possibly the most popular method for simulating columns today. Numerous studies have built upon this seminal work using a variety of systems and correlations (Hupen & Kenig, 2004; Klöker, et al., 2005).

While these techniques have been proven to accurately simulate complex columns to good accuracy, there has been little in the way of optimizing the design of packed absorption columns with regards to financial objective functions. With much work being done on complex reactive trayed columns, the relatively simple absorber optimization has often been overlooked. Mores, et al. (2012) used a deterministic NLP optimization of a packed column considering fixed packing type and size to optimize CO_2 absorption using monoethanolamine. They compared various methods and found that a simplified rate-based model was sufficiently accurate, with the inclusion of non-idealities shown to make little difference to the accuracy of the solutions. They solved the problem by dividing the column into sections and then calculated the non-equilibrium efficiencies in each stage. They found Onda's (1968) effective mass transfer area to be the most suitable. Their models showed that interesting and accurate designs can result from deterministic optimization strategies when applied to new systems. Their objective function was simplified as it considered CO_2 absorbed per total packing volume maximized.

The vast majorities of these studies were simulations where the height was solved for and either included only a fixed packing with the packing sizes decided on in advance, or with packing types totally ignored. Many also did not include flooding conditions as the

simulations had assumed diameters, and therefore velocities. In the problem presented in this paper, the packing sizes, column diameters, and flooding conditions are not known beforehand. These additional variables add major extra complexities. In addition to this, the more complex system of packed distillation columns with heat integration has often been the source of study for individual column optimization. In this study we are focused on the optimization of networks involving packed-bed gas-liquid absorbers only. These simpler systems can allow for the implementation of these extra packing and flooding characteristics as variables to allow for the accurate design of these systems without the use of fixed parameters.

The method to be used in this study entails an approach similar to that used by Short, et al. (2016a) for HENS, using an MINLP model with simplified equations for the network optimization, followed by a more rigorous optimization of the network's individual units. As opposed to Short, et al. (2016a), the new method of this paper, apart from it not being applied to HENS, but rather MENS, the individual unit optimization is done through an NLP optimization with the use of orthogonal collocation on finite elements (OCFE) in order to accurately represent the concentration profiles along the columns, as well as a novel costing model, that allows for the selection of optimal packing, and constraints on column length to diameter ratios and flooding considerations. The solutions of these rigorous individual column models are then used to "guide" the MINLP model in another iteration using correction factors, similar to the procedure utilized in Short et al. (2016a) for HENS. The methodology is described in detail below.

6.3 Methodology

This section will describe the methodology used in the examples. The methodology closely follows the methodology of Short, et al. (2016a), however it is applied to MENS, and not HENS, and includes a novel rigorous individual mass exchanger optimization using NLP optimization.

The network topology optimization closely resembles that used by other authors, with the SBS model of Azeez, et al. (2013) chosen as the basis for its consistent performance across a number of examples, and its relative simplicity. The reason for avoiding the more complex

model of Isafiade and Short (2016) is due to the authors' troubles in finding initial values without an initialization strategy due to the added complexities and non-linearities that easily lead to local optima, as well as the lack of a way to optimize the packed columns with regards to flooding and packing characteristics without the need for manual checking. The adjustments that have been made to this model are presented in the main body of the paper below, with the rest of the model equations presented in Appendix 6.A. The principal difference between the SBS model and the one used in this paper is the inclusion of correction factors. The full list of correction factors adopted are shown in Table 6.1. These correction factors, like those employed in Short, et al. (2016a, b), are included to allow for the simplified models in the MINLP section to converge upon the solutions obtained by the rigorous models used in the NLP sub-optimisation. The substantial difference between this model and the SBS is in the objective function. The objection function used in this study is shown in Equation 6.1.

$$min\left\{\begin{array}{c}\left[AF\left\{\sum_{r\epsilon R}\sum_{l\epsilon S}\sum_{k\epsilon K}\left(\begin{array}{c}\left[23805.\left(Dcor_{r,l,k}.D_{r,l,k}\right)^{0.57}.1{,}15.Hcor_{r,l,k}.H_{r,l,k}\right]+\\ \left[PackCost_{r,l,k}.\frac{\pi}{4}.\left(Dcor_{r,l,k}.D_{r,l,k}\right)^{2}.Hcor_{r,l,k}.H_{r,l,k}\right]\end{array}\right)\right\}\right]+\\ FC\left(\sum_{r\epsilon R}\sum_{l\epsilon S}\sum_{k\epsilon K}\left(z_{r,l,k}\right)\right)+\left[\sum_{l\,\epsilon\,S}AC_{l}\,.L_{l,p}\right]\end{array}\right\} \quad (6.1)$$

Where AF is the annualisation factor, $D_{r,l,k}$ is the diameter of the column, $H_{r,l,k}$ is the packed height of the column, $z_{r,l,k}$ is the binary variable associated with the existence of a match, and FC is the fixed cost/installation cost of an exchanger. The first term in the first square bracket is related to the annualized variable capital cost of the exchangers, where the 23805 value is related to the cost of the column shell, the value of 1.15 is to account for 15% inactive space in the column, thereby giving the true height of the column, as opposed to just the packed height. The second term, which includes the $PackCost_{r,l,k}$ variable, relates to the cost of the packing within the mass exchanger. AC_l is the cost of the lean streams or MSAs and $L_{l,p}$is the lean stream flowrate.

The other correction factors are included in Equation 6.2 below that are used to calculate the overall mass transfer coefficient.

$$kya_{r,l,k} = ky_{r,l,k}.kyCor_{r,l,k}.ai_{r,l,k}.aiCor_{r,l,k} \qquad k \in K, l \in L, r \in R \qquad (6.2)$$

Where $ky_{r,l,k}$ is the initial overall mass transfer coefficient which is later corrected by $kyCor_{r,l,k}$ between each iteration, $ai_{r,l,k}$ is the initial interfacial area of the packing which is later corrected by $aiCor_{r,l,k}$ between each iteration, and $kya_{r,l,k}$ is the combination of

the two. Note that in the MINLP all of these are parameters that are updated only after the NLP run; therefore not adding to the complexity of the MINLP model. Equation 6.2 is one of the equations used in the calculation of the height of each packed column, with its use in the determination of $H_{r,l,k}$ shown in Appendix 6.A (Equation 6.A12). It should be noted that the height is calculated through the use of an approximation for the log mean composition difference (LMCD). This is done in the same way as other authors (Szitkai, et al., 2006; Azeez, et al., 2012; Isafiade & Short, 2016). While the approximation (Equation 6.A11) has favorable numerical properties, discussed in detail in Shenoy and Fraser (2003), it has been shown to provide poor estimates of the LMCD under certain conditions.

Table 6.1: *List and purpose of corrections used in the study*

Correction Factor	***Purpose***
$kyCor_{r,l,k}$	Correction for the overall mass transfer coefficient, $ky_{r,l,k}$
$aiCor_{r,l,k}$	Correction for the interfacial area of the packing, $ai_{r,l,k}$
$Hcor_{r,l,k}$	Correction to column height, $H_{r,l,k}$
$Dcor_{r,l,k}$	Correction for column diameter, $D_{r,l,k}$
$PackCost_{r,l,k}$	The updated packing cost obtained from the NLP step

Another difference between the original SBS model and the one in this study is the inclusion of unequal composition mixing. This was not included in the original models as the additional non-convex terms result in added computational difficulty, however in this study it was decided to include them to get potentially better solutions, as well as to allow for more interesting interplay between the NLP and MINLP subsections. Equations 6.3 and 6.4 below show the added equations that enable this.

$$G_r = \sum_{l \in L} fr_{r,l,k} \qquad k \in K \quad r \in R \tag{6.3}$$

$$L_l = \sum_{r \in R} fl_{r,l,k} \qquad k \in K \quad l \in S \tag{6.4}$$

where $fr_{r,l,k}$ and $fl_{r,l,k}$ represent split flows for rich and lean streams respectively in the particular interval. The mass balances across each interval of the SBS are therefore also altered to include these new variables:

$$fr_{r,l,k} \times \left(y_{r,k} - yr_{r,l,k}\right) = M_{r,l,k} \qquad k \in K, l \in L, r \in R \tag{6.5}$$

$$fl_{r,l,k} \times \left(yl^{*}_{r,l,k} - y^{*}_{l,k+1}\right) = M_{r,l,k,p} \qquad k \in K, l \in L, r \in R \qquad (6.6)$$

Where $yr_{r,l,k}$ and $yl^{*}_{r,l,k}$ are the rich streams' and lean streams' compositions at the streams' exit from the column.

The remaining equations are shown in Appendix 6.A (Equations 6.A1-6.A12) and together form an MINLP model. This MINLP model uses a mostly linear formulation that assumes many of the design parameters are constant for all columns; diameters, mass transfer coefficients, and packing characteristics, as well as the fact that pressure drops are not considered. It also uses other simplified equations that may result in columns that are wholly different when designed using detailed models. Even with all of the simplifications in the MINLP model, there have been numerous authors that have reported problems in finding globally optimal solutions, with infeasible solutions and local optima common. In addition, many authors have underlined the fact that it may be necessary to use specific initialization and bounding strategies in order to find suitable networks. Due to these issues that result from the problems associated with modern MINLP solvers and the model formulation, the method of Short et al. (2016a) for HENS, is applied here. Once the solution to the MINLP is found, the resulting mass balances and exchanger matches are fed into the NLP suboptimisation, which is used to optimize the designs obtained using more detailed model simulation.

6.3.1 Non-linear Programming Step

Since a great amount of research exists on the simulation of continuously contacting packed columns, it would be relatively simple to apply any of the methods that were mentioned in the literature review section. As was discussed in Short et al. (2016a), the method chosen for the detailed model of individual exchangers is not of primary concern with the methodology as any process simulator or method that improves the accuracy of the MINLP model's design can be utilized. Any software or methodology could be utilized in this step, for example commercial package AspenPlus ™ from Aspen Technology, free simulators such as ChemSep ™, heuristic design approaches, or a detailed NLP optimization, as was used in this study. A process simulator can be used such as ASPEN, HYSIS, or CHEMSEP, where the user's interaction with the model can perhaps allow for designs that are more intuitive or related to the designer's experience. Since the literature on the deterministic solver

optimization of packed columns is fairly sparse, a unique model is developed in this paper that allows for the packing characteristics to be simultaneously solved for, along with the other column specifics. In addition, the model allows for the simultaneous optimization of multiple columns in a network synthesis. These are further novel contributions of this paper. Since the solutions from the MINLP involve isothermal columns, the columns of this study are also modelled as such. This allows for simplified equations with regards to the thermodynamic properties of the fluids involved. With these exclusions the model is expanded to consider many variables, including the flooding considerations, multiple columns at once, and packing characteristics with the packing type fixed, as well as other design constraints. The inclusion of temperature and detailed thermodynamics can be included through the use of a process simulator or another optimization model where fewer design considerations are taken into account. The use of an NLP optimization here is also further justified in that it allows for the iterative procedure to be automated easily within a single optimization environment, such as GAMS (General Algebraic Modelling System) with reasonable solution speeds.

After the MINLP model solution is found, the solutions for the mass balances, flowrates, and compositions obtained are used to set up the optimization of the individual mass exchangers using detailed equations. Due to the focus of this paper being a general approach for the optimization of networks of mass exchangers, the individual exchangers are optimized using a basic, general differential equation, where the mass flow, *M*, across a differential height element *z* is given by:

$$\frac{dM}{dz} = N_{total} \tag{6.7}$$

Where N_{total} is the total mass flux over the element of packing. Since the examples dealt with in MENS are typically non-reactive, contain single contaminants, and inert bulk streams, this equation will be sufficient if *M* is assumed to be the change in mass of the contaminant from the rich stream to the lean stream over the element dz.

Orthogonal collocation on finite elements, employing Radau polynomials, is applied to solve this differential equation for each of the columns that result from the MINLP in order to transform this problem into an NLP. This is appropriate since we have known boundary conditions for the differential equations at the ends of the exchangers. The height of each column is divided into a number of elements, with orthogonal collocation applied in each element. Since the height of the columns is to be optimized in the NLP, the height of the

column and therefore the height of each element is a variable. The equation is then rewritten:

$$\frac{L_{r,l,k}}{h_{r,l,k,jj}}\frac{dx_{r,l,k,tt}}{du} = Flux_{r,l,k,tt} \tag{6.8}$$

$$\frac{G_{r,l,k}}{h_{r,l,k,jj}}\frac{dy_{r,l,k,tt}}{du} = Flux_{r,l,k,tt} \tag{6.9}$$

Where the subscripts *r,l,k* represent the individual match or exchanger from the MINLP section, and the subscripts *ii* and *jj* represent the collocation points and the finite elements respectively where $jj \in 1,2,\dots,NE$ and $ii \in 1,2,\dots,NC$. Subscript tt is the total number of gridpoint over the entire domain, i.e. $tt \in 1,2,\dots,(NE(NC-1)+1)$. $h_{r,l,k,jj}$ is the variable that determines the height of the individual finite elements (FEs). And for element *e*:

$$u = (z - \sum_{jj=1}^{e-1} h_{r,l,k,jj})/h_{r,l,k,e} \tag{6.10}$$

where

$$h_{r,l,k,e} = \sum_{jj=1}^{e} h_{r,l,k,jj} - \sum_{jj=1}^{e-1} h_{r,l,k,jj} \tag{6.11}$$

When OC is applied:

$$\frac{L_{r,l,k}}{h_{r,l,k,jj}}\sum_{ii=1}^{NC} A_{ii,tt}x_{r,l,k,ii} = Flux_{r,l,k,tt} \tag{6.12}$$

$$\frac{G_{r,l,k}}{h_{r,l,k,jj}}\sum_{ii=1}^{NC} A_{ii,tt}y_{r,l,k,ii} = -Flux_{r,l,k,tt} \tag{6.13}$$

Where $A_{ii,tt}$ is dependent on the interpolating polynomial, in our case, Lagrange polynomials.

In general, Radau collocation points are preferred as they allow for constraints to be set at the end of each element and give more efficient stabilization with high index differential algebraic equations (Biegler, 2007). Through the use of Radau collocation points, continuity can be maintained between elements easily with the last collocation point from the previous boundary providing the starting point for the next element. The boundary conditions for the first and last element for the supply and target concentrations for each column are provided from the MINLP.

The $Flux_{r,l,k,ii,jj}$ is related to the other variables in the model by the following relationship:

$$Flux_{r,l,k,ii,jj} = ky_{r,l,k}.ai_{r,l,k}.A_{r,l,k}.\left(Cr_{r,l,k,ii,jj} - He.Cl_{r,l,k,ii,jj}\right) \qquad (6.14)$$

Where $ky_{r,l,k}$ is the mass transfer coefficient, $ai_{r,l,k}$ is the interfacial area of the packing, $A_{r,l,k}$ the internal area of the column, $Cr_{r,l,k,ii,jj}$ and $Cl_{r,l,k,ii,jj}$ are the rich and lean stream concentrations at each collocation point, *ii*, and finite element, *jj*, respectively. He is the Henry's law coefficient and is the only parameter in this equation while all others are variables. If more rigorous thermodynamics are required and the isothermal assumption is removed, this should also be a variable related to the temperature. Since there exists no way to rigorously determine the overall mass transfer coefficients in packed columns, numerous correlations have been proposed. Pratt's correlation (Leva, 1953) was the method used by Hallale and Fraser (2000b) and Isafiade and Short (2016) and has been used in this study to determine the overall mass transfer coefficient.

$$ky_{r,l,k} = \frac{G'_{r,l,k}}{\in_{r,l,k}}.a_G.\left(\frac{de_{r,l,k}.G'_{r,l,k}}{\in_{r,l,k}.\mu_{r,l,k}}\right)^{-0.25}.Sc^{-0.667}\omega e^{\beta L_p} \qquad (6.15)$$

Where $G'_{r,l,k}$ is the superficial velocity of the rich/gas stream, $\in_{r,l,k}$ is the voidage of the selected packing, $de_{r,l,k}$ is the equivalent diameter of the packing, $\mu_{r,l,k}$ is the viscosity of the gas stream, a_G and β are experimental constants, L_p is the liquid flowrate at the periphery of the packing, ω is a function of the wetted packed surface, Sc is the Schmidt number. The term $\omega e^{\beta L_p}$ has been considered to be 1 to get an estimate of the mass transfer coefficient from the available experimental data, as has been done by other authors (Hallale & Fraser (2000b) and Isafiade & Short, 2016). It is important to notice that $ky_{r,l,k}$ is thus a function of the chosen packing's characteristics as well as the $G'_{r,l,k}$, which is a function of the column's diameter.

$$G_{r,l,k} = A_{r,l,k}.G'_{r,l,k} \qquad (6.16)$$

$$L_{r,l,k} = A_{r,l,k}.L'_{r,l,k} \qquad (6.17)$$

Equations 6.16 and 6.17 relate the volumetric flowrate of each stream in each match, $G_{r,l,k}$ for the gas/rich stream and $L_{r,l,k}$ for the liquid/lean stream, to the superficial velocities and the column areas. Note that the $G_{r,l,k}$ and $L_{r,l,k}$ are parameters obtained from the MINLP optimization, while the others are variables.

$$A_{r,l,k} = \frac{\pi}{4}.(D_{r,l,k})^2 \qquad (6.18)$$

Equation 6.18 simply relates the $A_{r,l,k}$ to the diameter of the column, $D_{r,l,k}$.

$$H_{r,l,k} \leq 25 * D_{r,l,k} \qquad (6.19)$$

$$H_{r,l,k} \geq 2 * D_{r,l,k} \qquad (6.20)$$

Equations 6.19 and 6.20 are heuristic limits, suggested by Douglas (1988), which set the height-to-diameter ratio between 2 and 25. A problem with the methodology of Isafiade and Short (2016) was that these were not limited, resulting in very short, but wide columns. These are not realistic as they take up large amounts of floor space on a plant, and have low fluid velocities (and thus mass transfer) which result in taller columns in practice, and therefore suboptimal capital costs (Ibrahim, 2014). In the methodology to follow, for Example 6.1, these constraints were active, however in Example 6.2, the flowrates were such that constraints 6.19 and 6.20 were removed in order for the large flowrates and low exchanger requirements to yield feasible solutions with regards to flooding resulting from high velocities.

$$15 * de_{r,l,k} \geq D_{r,l,k} \qquad (6.21)$$

Similarly, there is a relationship between column diameter and packing size. While this depends on the packing size, it has been suggested that for Raschig rings, a ratio of upwards of 15 is preferred (Ibrahim, 2014). In order to avoid having binary variables or disjunctive programming in the NLP optimisation, discrete packing characteristic tables were fitted to curves and then these were used to model the packing characteristics as continuous variables. These curve fits are shown in Appendix 6.B, and while the R^2 values of between 0.85 and 0.995 for the curves suggest inaccuracy, they were deemed accurate enough for the purposes of this model that uses numerous empirical correlations. Since the example studied involved the use of Raschig rings, the data was taken from Perry's Chemical Engineering Handbook (2008) for Carbon Steel Raschig rings of various size.

$$PackFact_{r,l,k} = 2.0034.\left(de_{r,l,k}\right)^{-1.564} \qquad (6.22)$$

$$SA_{r,l,k} = 5.0147.\left(de_{r,l,k}\right)^{-0.978} \qquad (6.23)$$

$$\epsilon_{r,l,k} = 0.0569.\ln\left(de_{r,l,k}\right) + 0.9114 \qquad (6.24)$$

$$PackCost_{r,l,k} = 397431.\left(de_{r,l,k}\right)^2 - 53449.\left(de_{r,l,k}\right) + 2366.1 \qquad (6.25)$$

Where $PackFact_{r,l,k}$ is the packing factor (m^{-1}), $SA_{r,l,k}$ the surface area of the packing (m^2/m^3), and $PackCost_{r,l,k}$ is the cost of packing ($/$m^3$), where, in the example it was updated from the year 1990 to the year 2000 using CEPCI indices. The year 2000 was chosen to allow for easier comparison with other authors, where the costing data for the examples

were also sourced. In order to determine the interfacial area, $ai_{r,l,k}$, from the $SA_{r,l,k}$ of the packing, Equation 6.26 from Onda (1958) is used.

$$ai_{r,l,k} = SA_{r,l,k} * \left(1 - \exp\left(-1.45.\left(\frac{\sigma_c}{\sigma_l}\right)^{0.75}.\left(\frac{\rho_l.L'_{r,l,k}}{\mu_l.SA_{r,l,k}}\right)^{0.1}.\left(Fr_{r,l,k}\right)^{-0.05}.\left(We_{r,l,k}\right)^{0.2}\right)\right) \quad (6.26)$$

This equation was verified by Mores et al. (2012) to be the most accurate prediction of $ai_{r,l,k}$ when compared with other empirical correlations. Here ρ_l is the density of the lean stream, σ_c is the critical surface tension of the packing and σ_l the surface tension of the liquid. $Fr_{r,l,k}$ is the Froude number:

$$Fr_{r,l,k} = \frac{SA_{r,l,k}.(L'_{r,l,k})^2}{g} \quad (6.27)$$

Where g is the gravitational constant. $We_{r,l,k}$ is the Weber number:

$$We_{r,l,k} = \frac{\rho_l.(L'_{r,l,k})^2}{SA_{r,l,k} * \sigma_l} \quad (6.28)$$

The Reynolds' numbers of the lean and rich streams, represented by $ReL_{r,l,k}$ and $ReR_{r,l,k}$ respectively, are determined using equations 6.29 and 6.30 below:

$$ReL_{r,l,k} = \frac{\rho_l.L'_{r,l,k}}{\mu_l.ai_{r,l,k}} \quad (6.29)$$

$$ReR_{r,l,k} = \frac{\rho_r.G'_{r,l,k}}{\mu_r.ai_{r,l,k}} \quad (6.30)$$

These Reynolds' numbers are used in the determination of the actual pressure drop across the column, $Pdrop_{r,l,k}$. The most common method to determine the flooding in a packed column is through the use of the generalized pressure drop correlation (GDPC) (Sinnot, 2005). Isafiade and Short (2016) used this correlation in order to select different packings for packed columns in their model, however the selection was done outside of the optimization. For this study the method of Jamialahmadi, et al. (2005) was used in order to account for the pressure drop explicitly in the NLP:

$$Pdrop_{r,l,k} = (94 \cdot \left(\frac{ReL_{r,l,k}^{1.11}}{ReG_{r,l,k}^{1.8}} + 4.4\right) \cdot 6 \cdot \left(1 - \frac{\in_{r,l,k}}{de_{r,l,k} * \in_{r,l,k}^3}\right).\rho_r.{G'_{r,l,k}}^2 \quad (6.31)$$

The point at which flooding occurs can be calculated using the method of Kister and Gill (1991), converted to the appropriate units:

$$FloodPoint_{r,l,k} = \frac{249.089}{0.3048} * 0.12 * \left(0.3048 * PackFact_{r,l,k}\right)^{0.7} \quad (6.32)$$

With the following inequality constraint used to ensure that the optimal design has a pressure drop below the flooding point:

$$FloodPoint_{r,l,k} \geq Pdrop_{r,l,k} \tag{6.33}$$

Combining these equations into an NLP model allows for the optimization of the individual packed columns obtained from the network generated in the MINLP by using more rigorous models. These rigorous models take into account many aspects that are not included in the MINLP, such as flooding considerations, column diameter, packing size, flux changes along the column, variations in the overall mass transfer coefficients, etc. The information obtained from the NLP can then be used to guide the MINLP to more accurate solutions based on these rigorous designs.

6.3.2 Correction Factors and Iterative Procedure

After the initial MINLP is run and the resulting exchangers rigorously optimized in the subsequent NLP, correction factors are obtained in a manner similar to that of Short et al. (2016a). These correction factors are implemented in order to "guide" the shortcut models in the MINLP toward the solutions obtained in the NLP. Each correction factor is just a simple linear correction to the current parameters and variables and each is described in the subsequent paragraph and listed in Table 6.1.

In all other mathematical programming-based MENS approaches, apart from that of Isafiade and Short (2016), the diameter has been kept as a fixed parameter. As discussed previously, the approach of Isafiade and Short (2016) has numerous limitations, specifically in flooding and packing considerations and in the additional numerical complexity in the MINLP. This new approach maintains the diameter as a fixed parameter in the MINLP, but now allows for the parameter to be updated in subsequent iterations to resemble the diameter of rigorously optimized packed columns by updating the diameter via the $Dcor_{r,l,k}$ correction factor. It is calculated by dividing the diameter of the detailed packed column design obtained from the NLP by the fixed diameter in the MINLP. Similarly, the $kyCor_{r,l,k}$ and $aiCor_{r,l,k}$ correction factors are used to update the fixed $ky_{r,l,k}$ and $ai_{r,l,k}$ respectively in the MINLP to represent the more accurate $ky_{r,l,k}$ and $ai_{r,l,k}$ determined in the NLP.

The $PackCost_{r,l,k}$ correction is the cost determined from the NLP that is inputted for each individual "match" in the MINLP, as determined by the NLP. Notice that as with the other

correction factors discussed thus far in this section, the $PackCost_{r,l,k}$ is a variable in the NLP model but inputted as a parameter into the MINLP.

$Hcor_{r,l,k}$ is the only correction factor applied to a variable in the MINLP. This correction factor is used to correct the height, $H_{r,l,k}$, of the packed column in the MINLP to more accurately represent the actual column design in the NLP. In the MINLP the height is calculated using equation 6.A12 (Appendix 6.A). This equation uses Chen's approximation of the log mean composition difference (Equation 6.A11), which has been shown by other authors to consistently underestimate the actual log mean composition difference, with large errors under certain conditions. A detailed comparison can be found in Isafiade and Short (2016). Since the NLP uses the differential equation, Equations 6.8 and 6.9, to represent the mass balance across the exchanger, the approximated $H_{r,l,k}$ in the MINLP is updated to represent this more accurately determined $H_{r,l,k}$ from the NLP.

As was the case in Short et al. (2016a), between each run the amount of change that a correction factor can undergo between iterations is limited. This is so that the solution space is not too drastically altered between runs so that potential solutions are not omitted during the algorithm. In the case of the example presented in this study, the correction factors were limited to a change of no more than 5% between iterations.

An improvement in this work over Short, et al. (2016a) is that both the network optimisation and individual exchanger optimisation are simultaneous in nature. The exchangers are optimized, not with heuristics as in the latter work, but in a deterministic fashion, which lends itself easily to automation and can therefore be much faster than the design methods employed in the previous study relating to HENS.

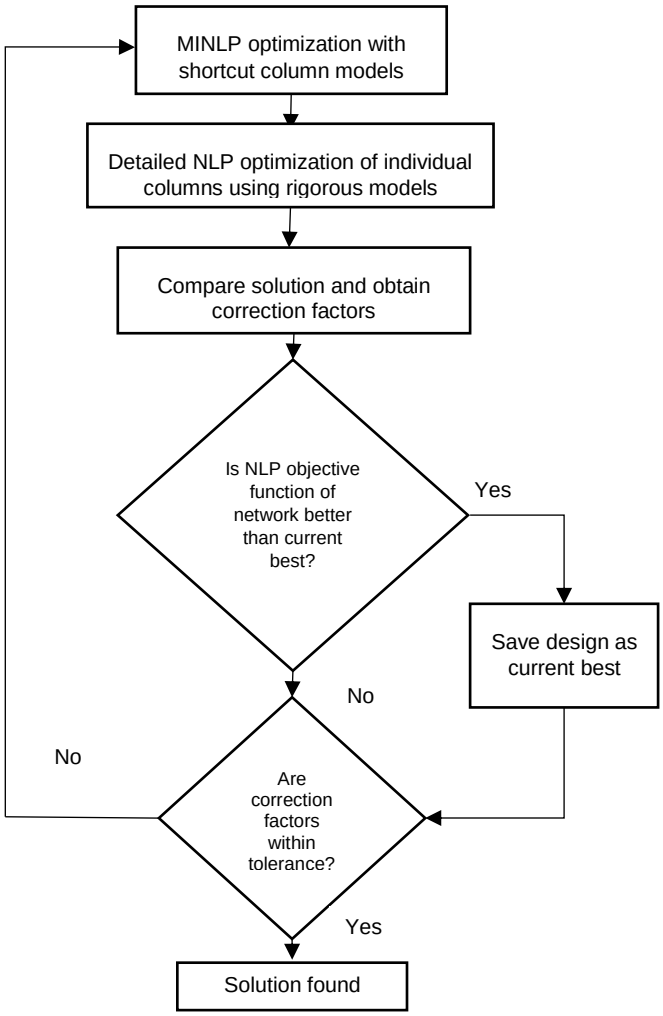

Figure 6.2: *Iterative procedure used in this study adapted from Short et al. (2016)*

6.3.3 Solution Strategy

This section will detail some of the methods used to ensure the model was robust enough to be automated, with special focus on the initialization strategy.

6.3.3.1 MINLP initialisation

When initializing the model it is important to have initialized parameters that underestimate the objective function in the MINLP step. This is done so that as the algorithm continues, no solutions are omitted (Short et al., 2016). The list of initializations for the parameters in the MINLP example are given in the text below. These are obtained by first running the MINLP and NLP once (with estimated parameters) in order to obtain the values that can be expected

and then using the values that are going to underestimate the objective function for all the parameters. For example, if the initial exploratory run gave $ky_{r,l,k}$ of 0.025, 0.03, 0.045, and 0.04 for the four exchangers, the initialization for all matches in the MINLP would be chosen as 0.05, as this would give an underestimation of the height and therefore objective function for all matches. Similar procedures were followed for the other parameters' initial values. The $Hcor_{r,l,k}$ initial value is 1, as this is only a correction to a variable that will be unknown.

6.3.3.1 NLP initialisation

Due to the size and non-convexity of the NLP subproblem described above, a specific initialization strategy was required. The initialization strategy involved a succession of NLP subproblems, each one more complicated and non-linear than the next, with the preceding subproblem providing the initialization for the next. In doing this, a potentially feasible solution was provided as a starting point for the subsequent more complex model until the final model, presented above was able to be solved. This strategy proved effective in providing feasible solutions to all of the subproblems and through all of the iterations of the examples, thus providing evidence for its robustness and efficacy.

6.3.4 Solution Algorithm/Solvers

The MINLP model shown in Appendix A is solved with DICOPT/GAMS with GAMS version 24.2.3 with CPLEX as the MILP solver, CONOPT as the NLP solver and DICOPT as the MINLP solver. The computer was equipped with an Intel Core™ i7-4700MQ 2.4 GHz CPU and 16 GB of RAM. The MINLP typically took between a few seconds to 4 minutes to solve, differing between successive iterations and examples based on the initialisations. The NLP made use of CONOPT/GAMS and typically took less than a second to solve, however if the initialisation subproblems are included, the solutions of all subproblems could sometimes run for up to 4 minutes.

The algorithm itself is easy to implement and does not add complexity to either the MINLP or NLP as it iterates. The NLP formulation proved to be extremely robust, finding feasible solutions for almost any starting network provided by the MINLP. The MINLP, however still had numerous issues finding feasible solutions throughout the iterative procedure, as reported by other authors. When the initial points were changed as a result of the correction factors from a previous iteration, the MINLP would often become infeasible, showing that

the issues around non-convex MINLPs described previously are well-founded and that the addition of further complexities at this level are not necessarily worthwhile. Whenever the procedure was halted due to an infeasibility in the MINLP, the value associated with the Big-M formulation was changed until a feasible solution was found and then the iterative procedure was resumed from that point.

6.4 Case Studies

The methodology was applied to two examples. The two examples are adapted from other authors, however there is little room for comparison as the costing in the objective functions and level of details included are far greater than that included by other authors. The first example is adapted from Isafiade and Short (2016), who originally adapted the problem from Hallale and Fraser (2000a) and Hallale and Fraser (2000b). The example includes two rich streams and two lean streams. One of the lean streams is a process MSA, limited in available flowrate, while the other is a more expensive external MSA with unlimited available flowrate. The problem data associated with this example is included in Table 6.2, with additional information included in Table 6.3. The second example is adapted from Hallale (1998) and consists of 5 rich streams and 3 lean streams, demonstrating the model's effectiveness at handling larger problems. The problem data for this problem are shown in Tables 6.4 and 6.5. For all problems considered, carbon steel Raschig rings were the only random packings considered and thus a_G=0.123 and σ_c = 0.075 for all examples (Perry et al., 2008). The SBS method was used in setting up the MINLP superstructure.

6.4.1 Examples

Table 6.2: *Stream data for Example 6.1. The AF was set to 0.2 and the FC = \$30,000. Sc = 0.7 for all liquid streams.*

Rich streams	G $(kg\ s^{-1})$	y^s	y^t	MSAs	L_c $(kg\ s^{-1})$	m	x^s	x^t	Cost $(\$\ yr^{-1})/(kg\ s^{-1})$
R_1	0.9	0.0700	0.0003	S_1	2.3	1.45	0.0006	0.0310	117,360
R_2	0.1	0.0510	.0001	S_2	∞	0.26	0.0002	0.0035	176,040

Table 6.3: *Other stream parameters used in Example 6.1*

Rich streams			
Match	**Density ($RHOG_{r,l}$)** $(kg\ m^{-3})$	**Viscosity ($\mu_{r,l}$)** $(Pa\ S)$	**Surface Tension (N.m^{-1})**
R_1,S_1	1.14	1.886×10^{-5}	0.0728
R_1,S_2	1.50	1.587×10^{-5}	0.0225
R_2,S_1	1.14	1.886×10^{-5}	0.0728
R_2,S_2	1.50	1.587×10^{-5}	0.0225
Lean streams			
S_1	900	.0011	
S_2	842.5	.0013	

Table 6.4: *Stream data for Example 6.2. The AF was set to 0.2 and the FC = \$15,000. Sc = 0.7 for all liquid streams.*

Rich streams	G $(kg\ s^{-1})$	y^s	y^t	**MSAs**	L_c $(kg\ s^{-1})$	m	x^s	x^t	**Cost** $(\$\ kg^{-1})$
R_1	2.0	0.005	0.001	S_1	1.8	1.2	0.0017	0.0071	0
R_2	4.0	0.005	0.0025	S_2	1.0	1.0	0.0025	0.0085	0
R_3	3.5	0.011	0.0025	S_3	∞	0.5	0.0	0.017	0.001
R_4	1.5	0.010	0.005						
R_5	0.5	0.008	0.0025						

Table 6.5: *Stream parameters for Example 6.2*

	Density ($RHOG_{r,l}$) ($kg.m^{-3}$)	**Viscosity ($\mu_{r,l}$) ($Pa.s$)**	**Surface Tension (N.m^{-1})**
All Rich streams	1.14	1.886×10^{-5}	
All lean streams	1000	0.001	0.0728

6.4.1.1 Example 6.1

The initial values for the MINLP for Example 6.1, prior to any correction factors being implemented were $ai_{r,l,k}$ = 300 m^2m^{-3}, $ky_{r,l,k}$ = 0.05 $kgs^{-1}m^{-3}$, $D_{r,l,k}$ = 0.35 m, and $PackCost_{r,l,k}$ = 1000 $\$m^{-3}$. The MINLP contains 175 equations, 10 discrete variables, and 161 continuous variables, with the NLP containing 21,107 equations and 21,130 variables. During the course of the algorithm, 2 distinct topologies were found, with various minor differences in split flows and mass transferred in each exchanger. During iterations 1 to 17, a solution network containing the same 5 columns was found. This solution (at iteration 17) is shown in Figure 6.3 with the detailed exchanger variables shown in Table 6.6. This 5-exchanger solution is very similar, in terms of exchanger mass duties, to the solution of Isafiade and Short (2016). While the solutions are similar in terms of mass flows in each exchanger, the associated costs differ with the solution found in Isafiade and Short (2016) which had a TAC of $371,275, while the rigorous solution presented here is $515,748. This is due, in part, to the addition of an exchanger fixed cost in this study, which is not present in Isafiade and Short (2016). If the additional fixed costs are removed from the objective function in this study, it would yield a solution of $365,748, even though the associated packing costs are higher in each of the exchangers than the packing used in Isafiade and Short (2016). This solution compares well to those of other authors and the current authors believe that this solution is a more accurate representation of the problem as the different sized packings' variable costs are considered, as well as a more accurate representation of the composition profiles along each column. This solution has been presented for comparison only, as the actual optimal solution was found after 27 iterations and had a different topology.

The optimal solution was found to be the network presented in Figure 6.4 and Table 6.7 with 4 units and a TAC of $485,273 ($365,273 if the fixed cost is removed). This network was found in the final iteration, after the MINLP and NLP solutions converged to within a percentage of each other, with very little change between the correction factors between runs 26 and 27. Figure 6.5 shows how the 2 solutions converged between runs 26 and 27 such that the key parameters present in the MINLP converge to the values found in the detailed NLP solution. It also clearly shows how the MINLP was able to find a better solution at run 18 as a result of the multiple starts and the inclusion of correction factors. It is also possible to note, though perhaps difficult to see, that the TAC from iterations 18 to 27 actually marginally improve. This is not a result of vastly different solutions to the NLP, but rather due to the fact that small changes are made in the MINLP as a result of the updated

correction factors included that guide the MINLP to slightly better values for the split flowrates and mass duties.

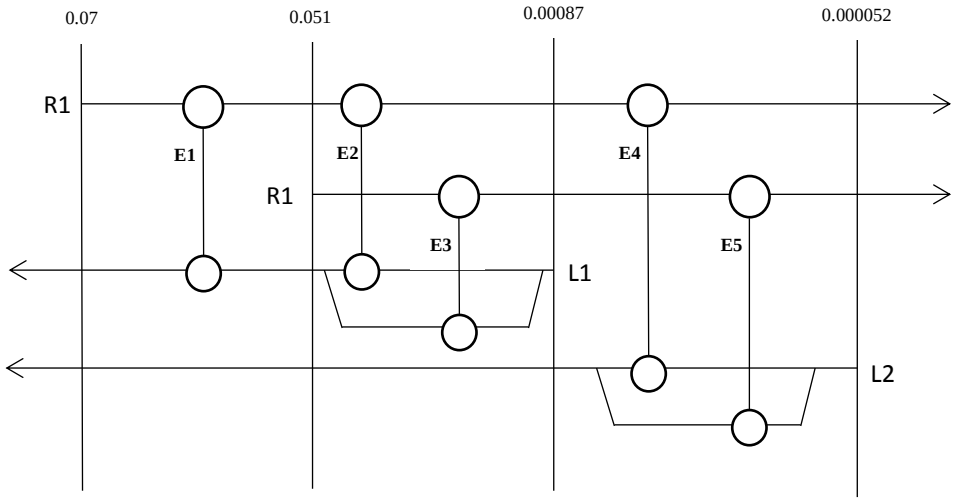

Figure 6.3: *Network topology for Example 6.1 for iteration 1-17*

Table 6.6: *Detailed exchanger designs for the 5-exchanger solution*

	E1	*E2*	*E3*	*E4*	*E5*
Packed Height (m)	1.543	4.532	1.391	2.369	1.460
Diameter (m)	0.729	0.725	0.418	0.664	0.321
Mass duty (kg/s)	0.04616	0.01597	0.004993	0.000583	9.65E-05
Rich flow (kg/s)	0.9	0.9	0.1	0.9	0.1
Lean flow (kg/s)	1.523	1.344	0.179	0.681	0.112
ky (kg/s/m^3)	0.049	0.049	0.026	0.045	0.032
Packing size (m)	0.036	0.036	0.021	0.033	0.016
Packing cost ($/m^3)	945.95	950.63	1423.27	1029.74	1610.17
ai (m^2/m^3)	127.91	128.57	219.55	140.156	285.10
Packing factor (m^{-1})	355.88	358.81	850.42	411.88	1282.12
Voidage	0.723	0.723	0.691	0.718	0.676
Pressure drop (kPa/m)	2.607	2.622	4.798	2.888	6.395

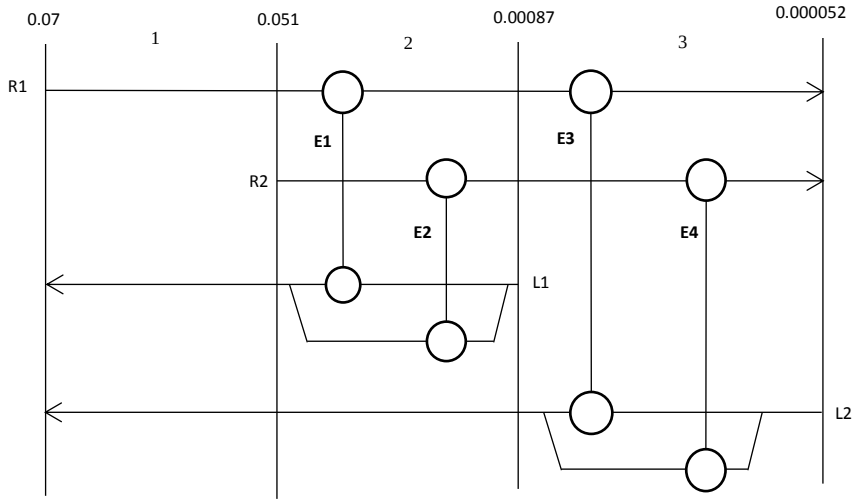

Figure 6.4: *Final network topology for Example 6.1*

Table 6.7: *Detailed exchanger designs for the optimal solution for Example 6.1*

	E1	*E2*	*E3*	*E4*
Packed Height (m)	5.801	1.472	2.327	1.411
Diameter (m)	0.725	0.413	0.664	0.322
Mass duty (kg/s)	0.06214	0.000589	0.004995	9.46E-05
Rich flow (kg/s)	0.9	0.9	0.1	0.1
Lean flow (kg/s)	1.351	0.685	0.172	0.112
ky *(kg/s/m³)*	0.049	0.026	0.045	0.032
Packing size (m)	0.036	0.021	0.033	0.016
Packing cost ($/m³)	950.43	1431.93	1029.66	1609.17
ai (m²/m³)	128.54	222.02	140.14	284.67
Packing factor (m^{-1})	258.686	865.57	411.83	1279.07
Voidage	0.723	0.691	0.718	0.676
Pressure drop (kPa/m)	2.622	4.858	2.888	6.385

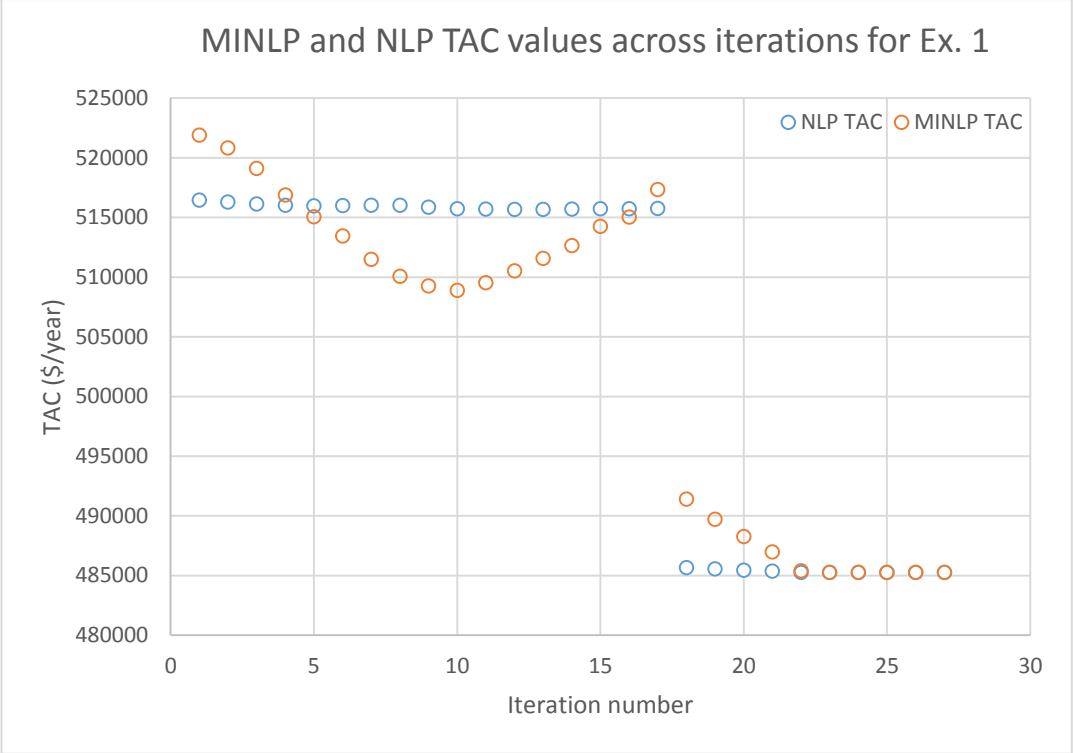

Figure 6.5: *Comparison of NLP TAC vs MINLP TAC across iterations for Example 6.1*

Table 6.8 shows the final values for the correction factors for each of the matches present. This table shows that it is difficult to predict the correct parameters in the MINLP without detailed knowledge of the problem beforehand, and that the degree to which they vary can be great, even when the same two streams are selected in different intervals. It also shows the large errors that can result from the simplified formulations used in traditional MINLP models when compared with more rigorous models.

Table 6.8: *Final correction factors for Example 6.1*

Match	$Hcor_{r,l,k}$	$Dcor_{r,l,k}$	$kyCor_{r,l,k}$	$aiCor_{r,l,k}$	$PackCost_{r,l,k}$
1.1.1	1.02947	2.0829	0.98	0.4264	0.94596
1.1.2	0.68949	2.071	0.98	0.4285	0.95043
1.2.3	1.00556	1.897	0.9	0.4672	1.02967
2.1.2	0.77612	1.18	0.52	0.7401	1.43193
2.2.3	1.02446	0.92	0.64	0.9489	1.60917

6.4.1.2 Example 6.2

Example 6.2 consists of five rich streams and three lean streams, with the corresponding stream parameters shown in Tables 6.4 and 6.5. This example is amongst the largest MENS problem found in current MINLP literature and therefore serves to demonstrate the method's efficacy at producing effective networks for larger problems. This example, first presented by Hallale (1998), has also been solved by Szitkai, et al. (2006), Emhamed, et al. (2007), Isafiade and Fraser (2008) and Azeez, et al. (2013). The initial values for the MINLP for Example 6.2 were $ai_{r,l,k}$ = 100 m^2m^{-3}, $ky_{r,l,k}$ = 0.05 $kgs^{-1}m^{-3}$, $D_{r,l,k}$ = 0.5m, and $PackCost_{r,l,k}$ = 550\$$m^{-3}$. The MINLP model contains 845 equations, with 63 discrete variables and 813 continuous variables and the NLP model contains 33,753 equations with 34,028 variables. Figure 6.6 shows the solution that was obtained for the initial MINLP. This solution, with 8 columns, gave a TAC of \$298,863, but when a more detailed solution was found by the NLP, a TAC of \$314,980 was found. The details and differences between the results for the first run are found in Table 6.9. This shows that, while the initial solution from the MINLP is good, it does not necessarily represent an achievable network in practice.

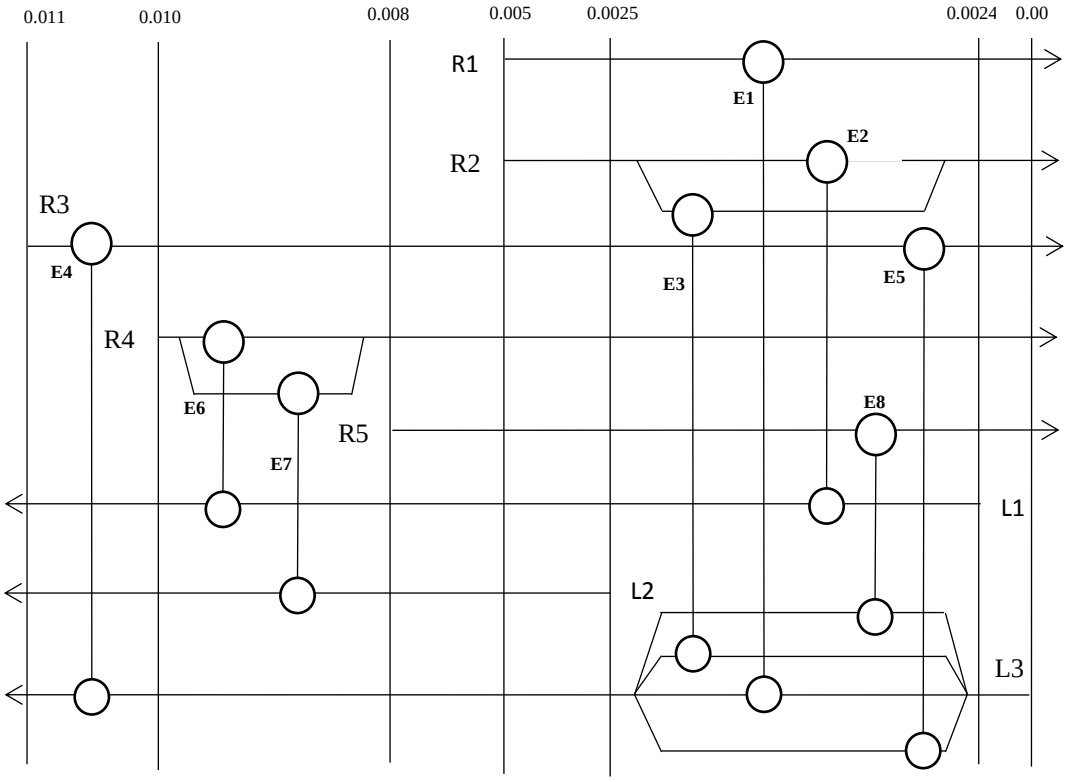

***Figure 6.6**: Initial MINLP network topology for Example 6.2 (Traditional solution)*

The algorithm was run for 100 iterations, generating many unique network configurations. Since the algorithm did not converge upon a solution during this time, it was decided to limit the number of iterations to 100 and to select the optimal solution as the network with the lowest TAC found in the NLP design. This network was found at iteration 35 and contained 8 packed columns with a TAC of $307,349. This detailed design is portrayed in Figure 6.7, with the column designs shown in Table 6.10.

***Table 6.9**: Initial solution network comparison of NLP to MINLP for Example 6.2*

	E1	***E2***	***E3***	***E4***	***E5***	***E6***	***E7***	***E8***
Mass duty (kg/s)	0.008	0.00338	0.00662	0.02036	0.00939	0.0051	0.0024	0.00275
Rich flow (kg/s)	1.791	1.309	1.432	5.543	1.945	1.3087	0.4	0.3759
Lean flow (kg/s)	2	1.79474	2.20526	3.5	3.5	1.1144	0.38563	0.5
Height (m):								
MINLP	4.306	2.0272	2.74	7.5111	3.463	1.8427	0.6921	0.7866
NLP	3.372	1.653	2.04	4.65	2.14	1.83	1.09	1.083
Diameter (m):								
MINLP	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
NLP	1.01	0.96	1.05	1.3	1.29	0.78	0.51	0.553
ky *(kg/s/m³):*								
MINLP	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
NLP	0.048	0.048	0.048	0.046	0.046	0.049	0.047	0.05
Packing cost ($/m³):								
MINLP	550	550	550	550	550	550	550	550
NLP	619.7	647.3	600.5	578.9	577.7	794.0	1167.3	1099.6
ai (m²/m³)								
MINLP	100	100	100	100	100	100	100	100
NLP	84.1	88.35	80.74	65.54	65.81	107.71	163.01	151.26
Packing size (m)	0.056	0.053	0.058	0.072	0.072	0.043	0.028	0.031
Pressure drop (kPa/m)	1.63	1.723	1.558	1.233	1.239	2.151	3.421	3.146

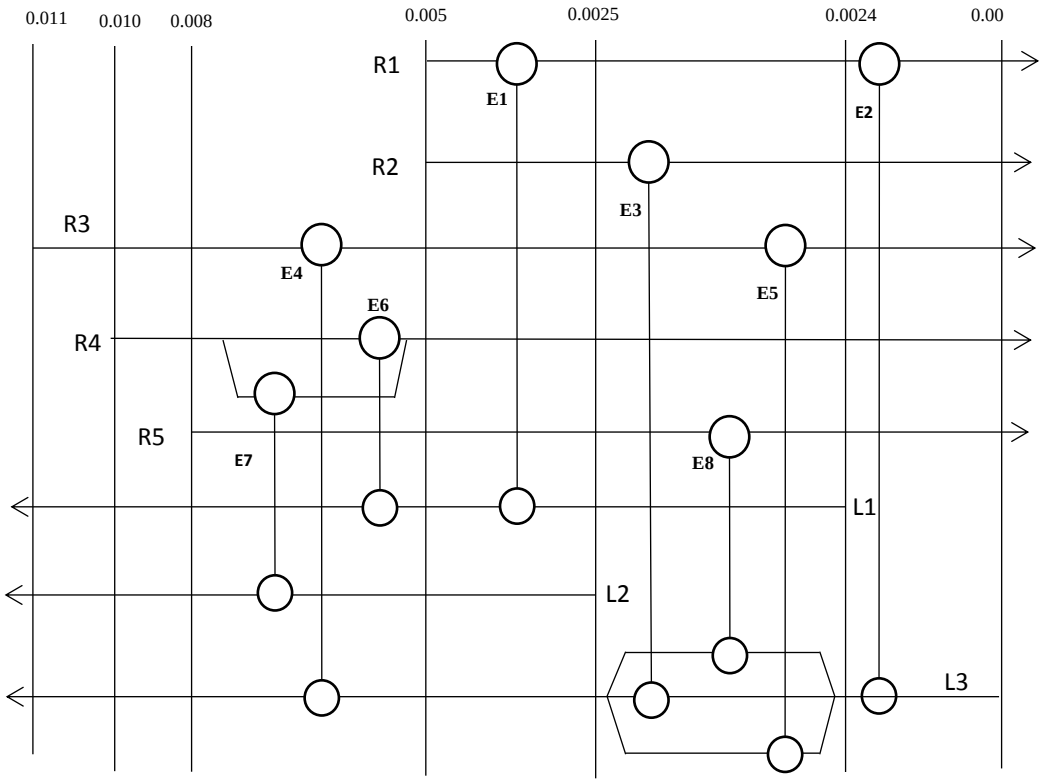

Figure 6.7: Optimal network topology for Example 6.2

Table 6.10: Optimal network column details for Example 6.2

	E1	***E2***	***E3***	***E4***	***E5***	***E6***	***E7***	***E8***
Packed Height (m)	1.289	0.913	2.78	4.21	2.78	1.78	1.09	1.19
Diameter (m)	1.01	1.025	1.376	1.3	1.3	0.782	0.512	0.558
Mass duty (kg/s)	0.00327	0.00473	0.01	0.0196	0.01012	0.0051	0.0024	0.00275
ky (kg/s/m³)	0.048	0.046	0.046	0.046	0.046	0.049	0.047	0.049
Packing size (m)	0.056	0.057	0.076	0.072	0.072	0.043	0.028	0.031
Packing cost ($/m³)	620.86	611.03	602.7	578.91	577.90	794.24	1167.3	1091.9
ai (m²/m³)	84.3	82.65	61.99	65.536	65.77	107.74	163.00	149.99
Packing size (m)	0.056	0.057	0.076	0.072	0.072	0.043	0.028	0.031
Rich flow (kg/s)	1.2921	5.5561	2.6445	5.5561	2.46611	1.2921	0.4	0.4455
Lean flow (kg/s)	2	2	4	3.5	3.5	1.1143	0.3857	0.5
Pressure drop (kPa/m)	1.635	1.599	1.159	1.233	1.238	2.152	3.421	3.116

The large differences between the fixed parameters in the MINLP and NLP solution are corrected through correction factors in the subsequent iterations. Figure 6.8 shows the TAC values of the NLP and MINLP over the entire algorithm and Figure 6.9 shows the relative differences. It is clear from these two figures that, as the algorithm progresses, the solutions for the networks more closely represent each other, with the final 10 iterations producing differences between the MINLP and NLP of less than 2% between iterations. While these solutions allow for the MINLP to select networks that can be created in reality, these solutions are not very close to the optimal solution.

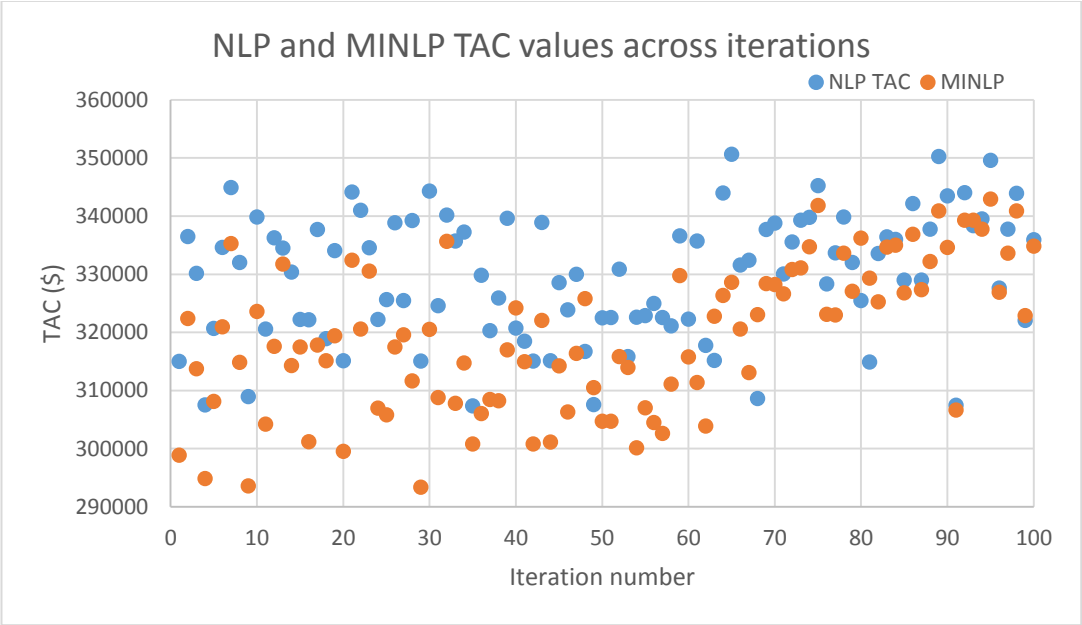

Figure 6.8: *Comparison of NLP solution to MINLP solution over the iterations*

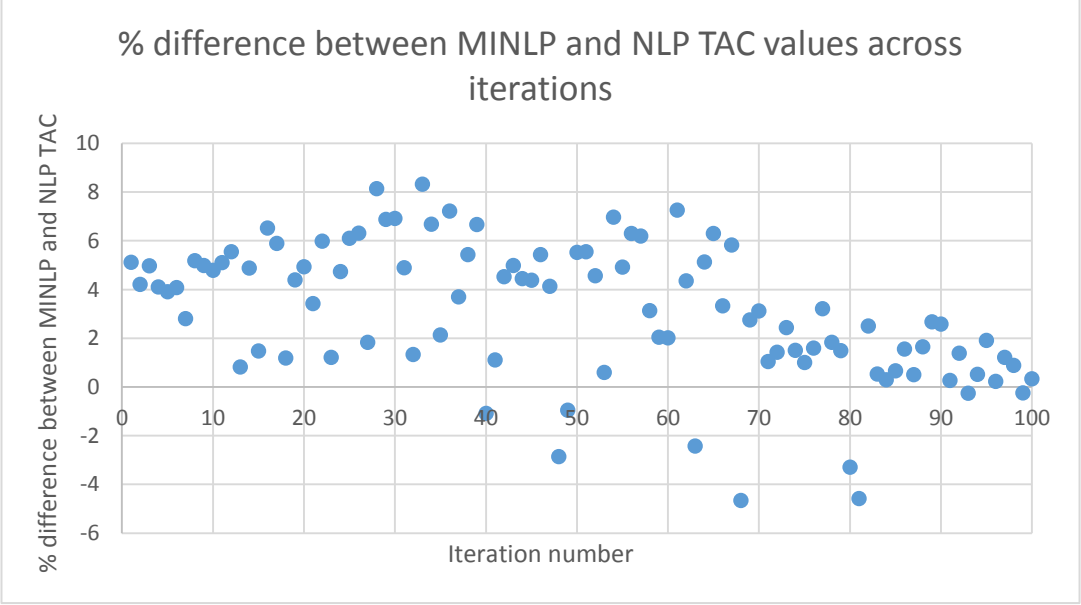

Figure 6.9: *Relative difference between NLP solutions to MINLP solutions over the iterations*

The generation of so many different candidate networks is advantageous in finding a global optimal network in such a large non-convex system. While it is impossible to guarantee that it is a globally optimal solution, due to the fact that deterministic solvers cannot be shown to guarantee globally optimal solutions in non-convex systems of equations, the multi-start procedure and the many different initial points provided by the shifting correction factors during each iteration mean that the chances that the solution is globally optimal is much higher. It can be noted from Figure 6.8 that a number of the NLP TACs are very close to the optimal solution (iterations 4, 35, 49, and 91). Upon detailed analysis of these networks it can be found that these are 4 distinct networks with very close TACs, but differing topologies. This means that it is very difficult for the MINLP solvers to find globally optimal solutions as multiple discrete variables give similar TACs. Through the use of this methodology, this problem is helped and shows that for large problems the methodology presented in this study can be particularly effective.

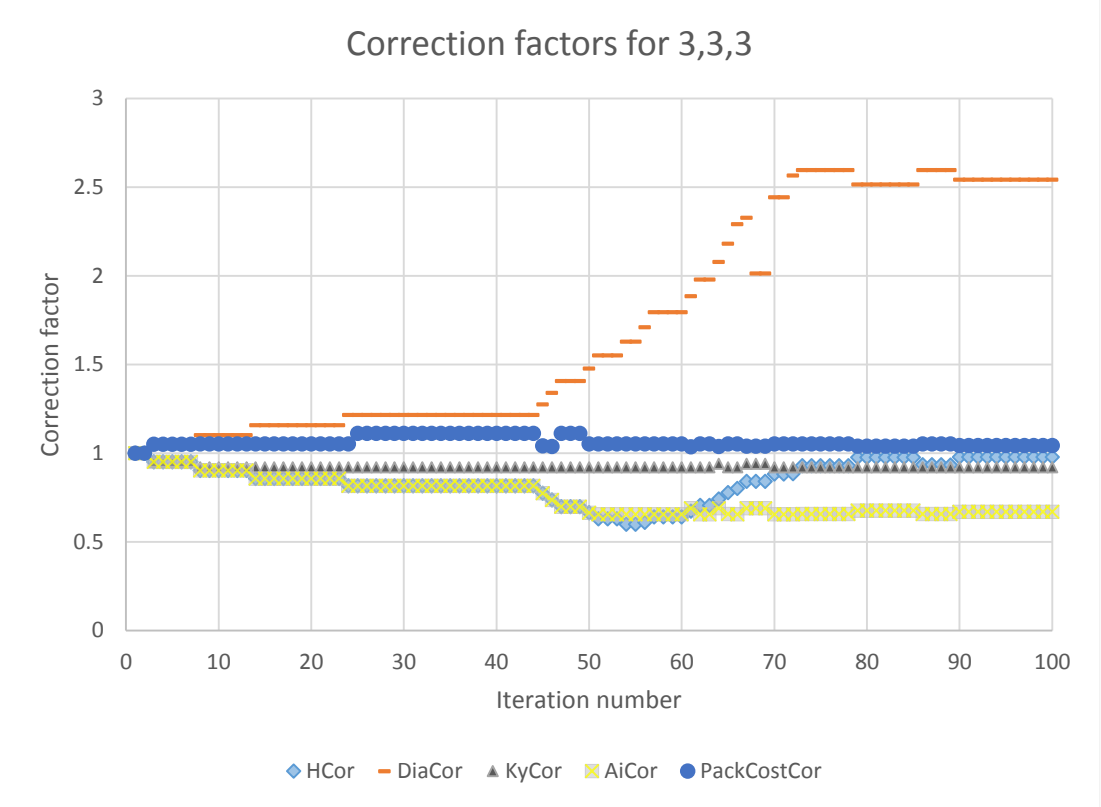

Figure 6.10: *Correction factors for a specific match (rich stream 3 with lean stream 3 in interval 3) over the iterations of the algorithm.*

Figure 6.10 acts as a visual demonstration of the way in which correction factors change over the course of the algorithm. The long periods where the correction factor is unchanged is due to the fact that the specific match (i.e. 3,3,3) is not chosen during those iterations, or

that there is no difference between the parameter in the MINLP and the correlating variable in the NLP. The figure shows the way in which the change to the correction factor is limited to not more than a 5% change between iterations. The full list of correction factors at the respective values at iteration 100 are included in Appendix 6C. An important question to raise here is why the MINLP generates such vastly different networks between certain iterations, despite the fact that the changes to the parameters are of such a small scale between the iterations. The reason for this is due, most likely, to the fact that many of the solutions generated by the MINLP are actually locally optimal.

It must be noted that the generation of the individual columns at the NLP level were found fairly quickly, with no iterations terminating due to infeasibilities. This demonstrates the robustness of the NLP formulation, as well as the abilities of CONOPT. However, this cannot be said of the MINLP. The solution procedure was interrupted numerous times, especially for Example 6.2, due to DICOPT's inability to find feasible solutions for the MINLP based on the new starting points provided by the changing correction factors. The non-convexities in the MINLP and larger numbers of binary variables in this example can be found to be the reasons for the solvers' poor performance. While it may be possible to use the "fairly linear" MINLP model of Szitkai et al. (2006) to allow for more robustness in the MINLP, it was decided to use a non-iso compositional interval based superstructure model to avoid the exclusion of potential networks. The 'restarts' added substantially to the total time of solution for the method, as at each restart, new values for the value of M in the "big-M" constraint would help the solver to find feasible solutions. It is suggested that in order to increase the robustness and reduce the time for the solution of the method that either a convex formulation for the MINLP be found and used, or that improvements to the MINLP solver technology be made.

Every column in every iteration for both of the examples studied had pressure drops that were at the bounds set by the constraint in Equation 6.33. This shows that the constraining factor in the selection of the packing size is, as other authors have shown, the point at which flooding occurs. The inclusion of a variable cost that includes the fact that smaller packing sizes are more expensive barely impacts the selection of smaller packing sizes, meanwhile the trade-off between diameter and packing size is vitally important in the design of an optimally priced column. In previous methods the trade-off between the costs of internals, packing sizes, and diameters have not been considered, with other authors (Hallale & Fraser, 2000b; Isafiade & Short, 2016) using fixed packing parameters and subsequently changing these fixed parameters in the following run after flooding was considered in post processing.

In addition, this study has presented a way to include these trade-offs implicitly in the network generation stage, while not adding further complexities associated with non-convexity into the MINLP. A further comment is that it can be seen that L/D constraints (Equations 6.19 and 6.20) prove to have little effect on the optimal solutions, as the constraints on packing size, diameter, and pressure drop are the actual limiting factors.

6.5 Conclusions

In this study mass exchange networks were successfully optimized and designed in detail using a novel combination of MINLP topology optimization based on shortcut models paired with a rigorous NLP individual unit optimization. This is a successful extension of the work of Short, et al. (2016a), for heat exchanger network synthesis, to mass exchanger network synthesis. The novel 2-step synthesis model allows for the use of relatively simple models in the network synthesis, allowing for fewer non-convexities and more robust solutions, while still accommodating details of the design that affect the capital costs of the final design from rigorous NLP model, such as changes to the overall mass transfer coefficient, column diameters, costs associated with different sizes of packing, flooding considerations, as well as an overall height correction factor to account for reported discrepancies in the log mean composition difference approximation.

The model works by first deriving the network using an MINLP optimization method that utilizes shortcut models for the synthesis of the individual heat exchangers, with fixed mass transfer coefficients, packing costs, and column diameters, as well as using a log mean composition difference approximation, as has been done by other authors. The solution, with mass balances fixed, is then sent to an individual mass exchanger optimization that is modelled as an NLP utilizing a more rigorous approach including a differential equation that is converted into algebraic form using orthogonal collocation, common methods for determining the overall mass transfer coefficients, as well as newly developed equations for determining the optimal packing size based on flooding considerations and detailed costing functions. Once the optimal exchangers are found in this rigorous method, the shortcut models in the MINLP are updated with a series of correction factors. These correction factors are used to more accurately represent rigorous solutions in the shortcut models, while not

increasing the level of non-linearity within the MINLP. The MINLP is then re-run and new designs obtained and the process repeated until the model converges on a solution where the NLP objective function is identical to the MINLP objective function, or until a maximum number of iterations is reached. During each iteration the current best solution, based on the total annual cost of the rigorously designed (NLP) network, is saved and compared, meaning that the converged solution does not necessarily have to be the optimal network. The correction factors are limited to a maximum of 5% change between iterations to ensure that potentially optimal solutions are not omitted.

Previous methods of MENS have reported great difficulty in finding feasible solutions, especially in large problems, even with the relatively simplified formulations used. This is due to the fact that modern MINLP solvers are imperfect and fail to guarantee a global optima. This method allows for the MINLP model to remain simple, while still accounting for the intricacies involved in the design and optimization of mass exchanger columns using rigorous models. The rigorous NLP optimization in the outer loop gives realistic columns that can be used in a real plant scenario, whereas the models of many other authors have given designs that have not been verified and were often unrealistic in size and therefore not suitable for industrial application.

While the novel method cannot guarantee globally optimal solutions, the multiple iterations with a variety of starting conditions as a result of the correction factors means that it is much more likely than other methods at finding the global optimum and this has been demonstrated in the examples, where new topologies were found after a number of iterations that gave superior total annual costs.

Further work is planned on applying the principles derived in this work with different NLP column optimization methods, water networks, and combined mass and heat exchanger networks that can include rigorous thermodynamic models for the heat and mass transfer coefficients along the column.

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6.7 Acknowledgements

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6.A Appendix 6A: MINLP Model Equations

The MINLP is modelled in similar fashion to the existing MENS MINLP models discussed in the body of the paper. The basis for the model is that of Azeez, et al. (2013), with an updated objective function, as well as the additional equations 6.1 to 6.66 that ensure that iso-compositional mixing is not enforced. These equations and the explanation are found in the main body of the study. In this formulation, the superstructure is defined by the supply compositions of the rich and lean streams, as illustrated in Figure 6.1. The following paragraphs show the equations used in the MINLP optimization section of this study.

If we use Figure 6.1 as an illustrative example, the following equations will represent the superstructure in the model; where the compositions are sorted so that the highest supply composition is on the left of the superstructure and compositions monotonically decrease towards the right:

$$\begin{aligned}
&\text{k = 1:} \quad Y^{S}_{R1,1} = y_{R1,1}; \quad Y^{*t}_{L1,1} = y^{*}_{L1,1}; \quad Y^{*t}_{L2,1} = y^{*}_{L2,1} \\
&\text{k = 2:} \quad Y^{S}_{R2,2} = y_{R2,2} \\
&\text{k = 3:} \quad Y^{*S}_{L2,3} = y^{*}_{L2,3} \\
&\text{k = 4:} \quad Y^{*S}_{L1,4} = y^{*}_{L1,4}; \quad Y^{t}_{R1,4} = y_{R1,4}; \quad Y^{t}_{R2,4} = y_{R2,4} \qquad (6.\text{A1})
\end{aligned}$$

Equations 6.A2 and 6.A3 ensure that compositions decrease monotonically along the superstructure.

$$y_{r,k} \geq y_{r,k+1} \qquad k \in K, \qquad r \in R \qquad (6.\text{A2})$$

$$y^{*}_{l,k} \geq y^{*}_{l,k+1} \qquad k \in K, \; l \in S, \qquad (6.\text{A3})$$

Equations 6.A4 and 6.A5 represent the overall mass balances for all streams across all of the intervals. These equations guarantee that the target compositions are met.

$$(Y^{S}_{r} - Y^{t}_{r})G_r = \sum_{k \in K} \sum_{l \in S} M_{r,l,k} \qquad r \in R \qquad (6.\text{A4})$$

$$(Y^{*t}_{l} - Y^{*S}_{l})L_l = \sum_{k \in K} \sum_{r \in R} M_{r,l,k} \qquad l \in S \qquad (6.\text{A5})$$

Where the supply and target compositions of the rich streams are represented by Y^{S}_{r} and Y^{t}_{r} respectively, while the supply and target compositions of the lean streams are represented by Y^{*t}_{l} and Y^{*S}_{l} respectively. $M_{r,l,k}$ is the amount of mass exchanged between the rich stream, r, and lean stream, l, in interval, k. G_r and L_l are the flowrates of the respective rich and lean streams.

Similarly to equations 6.A4 and 6.A5, each rich and lean stream requires a mass balance over each interval of the superstructure. Equations 6.A6 and 6.A7, below, represent the rich and lean streams respectively.

$$\left(y_{r,k} - y_{r,k+1}\right)G_r = \sum_{l \in S} M_{r,l,k} \qquad r \in R \quad k \in K \qquad (6.\text{A6})$$

$$\left(y^{*}_{l,k} - y^{*}_{l,k+1}\right)L_l = \sum_{r \in R} M_{r,l,k} \qquad l \in S \quad k \in K \qquad (6.\text{A7})$$

Where the intermediate compositions of rich streams are represented by $y_{r,k}$ and those of the lean streams by $y^*_{l,k}$. In order to ensure numerical stability Big-M constraints (Equation 6.A8) are included that force the mass exchanged in an exchanger to zero when the binary variable $z_{r,l,k}$ takes a zero value (Szitkai, et al., 2006).

$$M_{r,l,k} - \Omega_{r,l} z_{r,l,k} \leq 0 \quad r \in R, l \in S, k \in K \qquad (6.A8)$$

Where the scalar $\Omega_{r,l}$ is an upper bound on the amount of mass that can be exchanged between the two streams in question. Equations 6.A9 and 6.A10 are utilized to calculate the driving forces at the ends of each packed column, represented by $dy_{r,l,k}$.

$$dy_{r,l,k} \leq y_{r,k} - y^*_{l,k} + \Phi_{r,l}\left(1 - z_{r,l,k}\right) \quad k \in K,\ r \in R,\ l \in S \qquad (6.A9)$$

$$dy_{r,l,k+1} \leq y_{r,k+1} - y^*_{l,k+1} + \Phi_{r,l}\left(1 - z_{r,l,k}\right)\ k \in K, r \in R,\ l \in S \qquad (6.A10)$$

In a similar formulation to 6.A8, when a binary variable has a value of '1' then the driving force, $dy_{r,l,k}$, is calculated. In order to avoid the possibility of negative driving forces, the parameter $\Phi_{r,l}$ is included. This parameter represents the upper bound of the driving force and is calculated from the problem-specific stream data.

Due to the presence of logarithms in the actual LMCD equation singularities may form, resulting in solver failure. In order to circumvent these issues, various approximations have been posited. A review of the various approximations is available in Shenoy and Fraser (2003). This study makes use of the most common approximation, that of Chen (1987) (6.A11).

$$LMCD_{r,l,k} = \left[\frac{\left(dy_{r,l,k}\right)\left(dy_{r,l,k+1}\right)\left(dy_{r,l,k} + dy_{r,l,k+1}\right)}{2}\right]^{\frac{1}{3}} \qquad (6.A11)$$

The height of each column (or exchanger) is calculated using equation 6.A12. Note that in this equation, the LMCD is approximated and the mass transfer coefficient, $kya_{r,l,k}$, column diameter, $D_{r,l,k}$, and column diameter correction factor, $Dcor_{r,l,k}$, are not variables in the MINLP optimization and are thus fixed during each successive MINLP run.

$$H_{r,l,k} = \frac{M_{r,l,k}}{kya_{r,l,k} . \frac{\pi}{4} (Dcor_{r,l,k} . D_{r,l,k})^2 \times LMCD_{r,l,k}} \qquad (6.A12)$$

It should be noted that $kya_{r,l,k}$ is the mass transfer coefficient multiplied by the interfacial area, described by Equation 6.2 in the main body of the paper and that the diameter is modified by a correction factor, with the details of its use described fully in the main body of the paper.

Equations 6.A1-6.A12, combined with Equations 6.1-6.6, presented in the body of the text, represent the full MINLP model that is utilized in the paper. The implementation of this MINLP with the NLP suboptimisation is detailed in the main paper.

6.B. Appendix 6B: Packing Characteristics Equations

As mentioned in the main body of the paper, the packing characteristics are needed in the NLP section of the model in order to allow for an optimal packing size to be selected. The packing characteristics are taken from standard tables found in Perry's Chemical Engineering Handbook (2008) and fitted to curves. The figures 6.B1-6.B5 show the curves, as well as the equations used in the model with their R^2 values. While the fits are in no way ideal, the authors feel that they are within a good enough tolerance for the purposes of the model. The tables below are for carbon steel Raschig rings, as done by other authors for the example studied. The costing, seen in Figure 6.B4, has been updated from the original date of 1990 to 2000 in order to compare with the date of the other costing functions used by other authors.

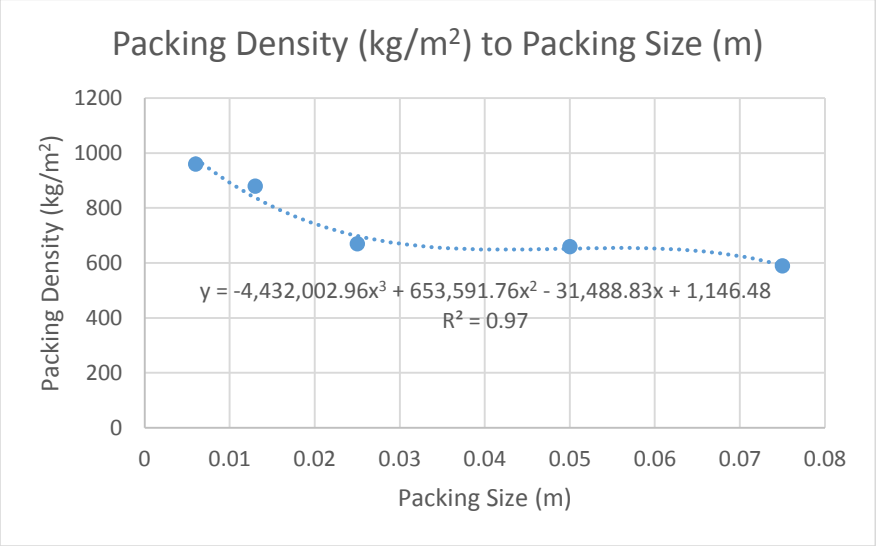

Figure 6.B1: *Curve fit of data from Perry's Chemical Engineering Handbook (2008) for Packing Density versus Packing Size*

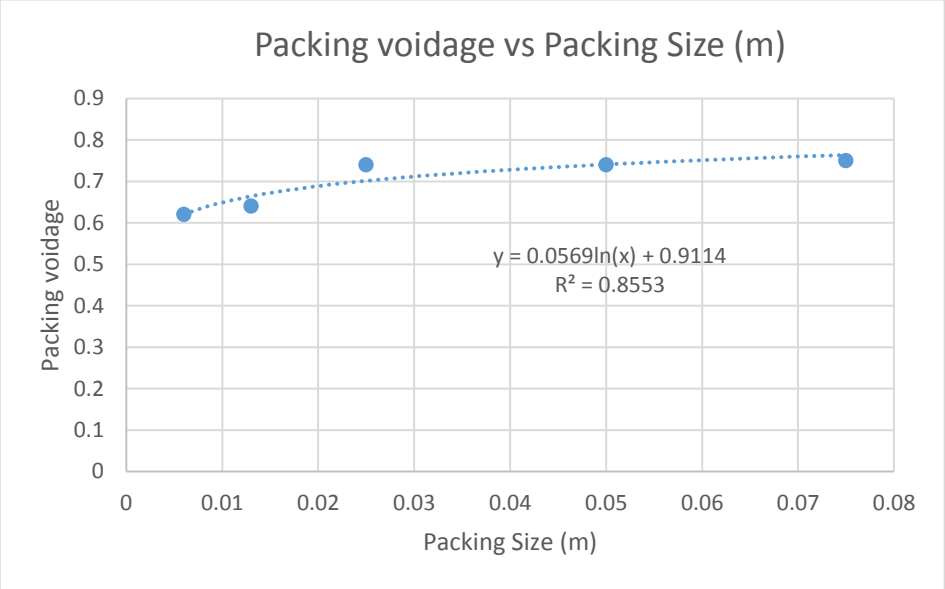

Figure 6.B2: *Curve fit of data from Perry's Chemical Engineering Handbook (2008) for Packing voidage versus Packing Size*

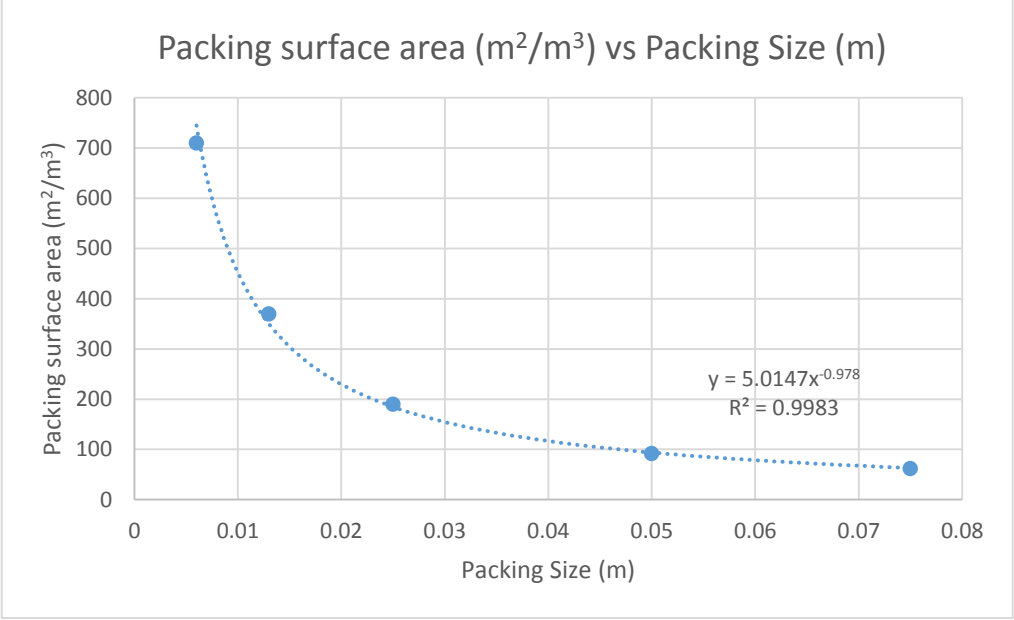

Figure 6.B3: *Curve fit of data from Perry's Chemical Engineering Handbook (2008) for Packing surface area versus Packing Size*

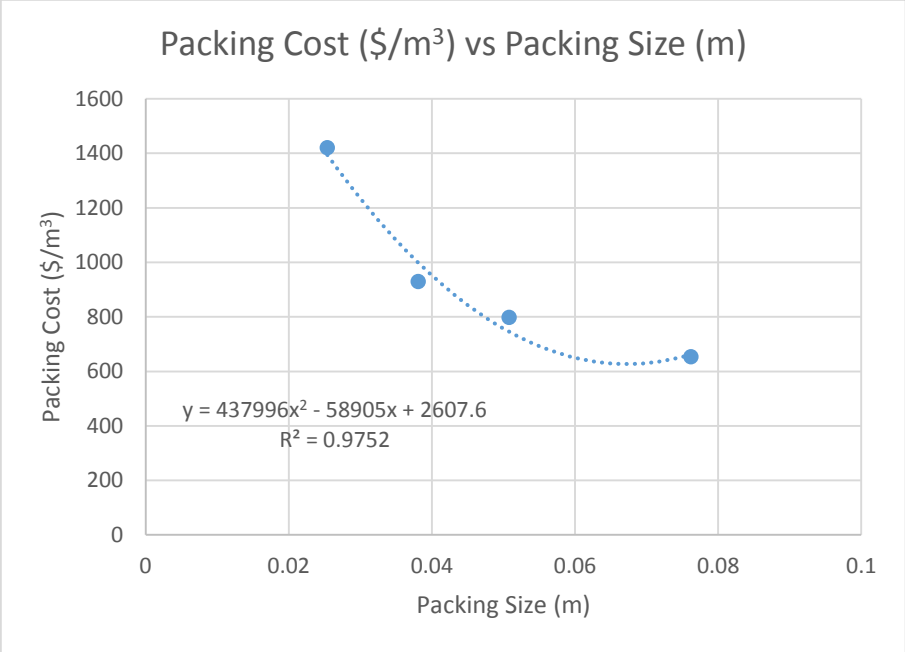

Figure 6.B4: *Curve fit of data from Perry's Chemical Engineering Handbook (2008) for Packing costing vs Packing Size*

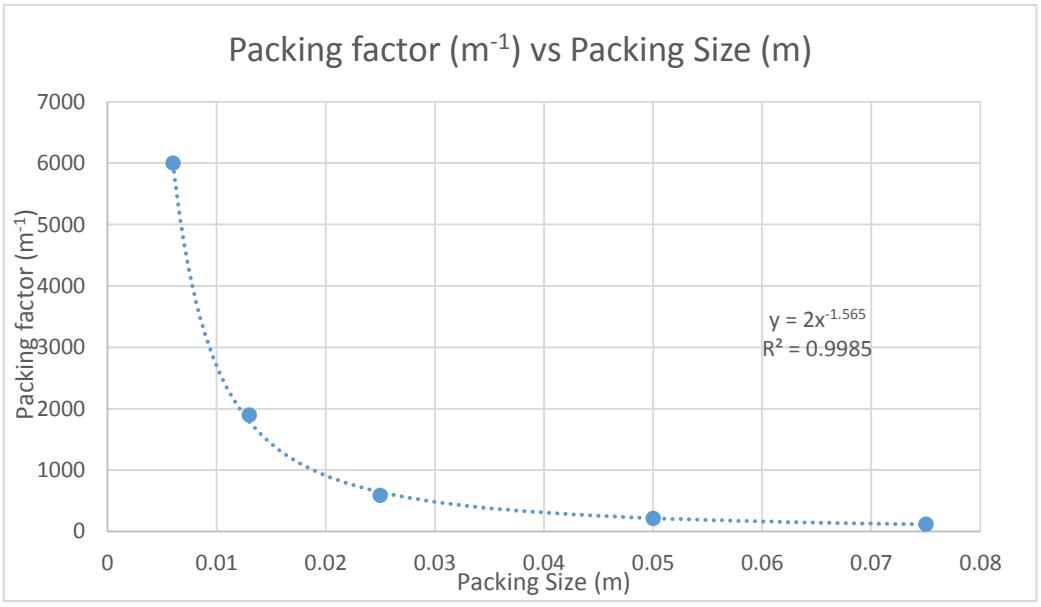

Figure 6.B5: *Curve fit of data from Perry's Chemical Engineering Handbook (2008) for Packing factor vs Packing size*

6.C. Appendix 6C: Final Correction Factors for Example 6.2

This appendix contains the final values for the correction factors used in Example 6.2 (i.e. at iteration 100). A value of 1 means that the match was not selected at any point during the algorithm. It is shown as it gives the reader an idea of the varying and unpredictable nature of the corrections that are required and how the simplified equations in standard MINLP methods are not able to accurately predict the actual sizes and internals of packed columns without access to more detailed equations.

Table 6.C1: *Values for the correction factors at iteration 100 for Example 6.2*

	HeightCor	*DiaCor*	*KwCor*	*AiCor*	*PackCostCor*
1.1.4	1.07782	1.795856	0.96	0.8422	1.1280
1.1.5	1.02659	1.710339	0.96	0.8429	1.1287
1.1.6	1	1	1	1	1
1.2.4	1.20728	2.002	0.96	0.8461	1.1323
1.2.5	1	1	1	1	1
1.2.6	1	1	1	1	1
1.3.4	0.95	1.05	0.95	0.95	1.05
1.3.5	1.2224	2.016	0.96	0.8407	1.1263
1.3.6	1.2732	2.014	0.96	0.8413	1.1270
2.1.4	0.9429	1.4071	0.94	0.9841	1.2181
2.1.5	0.86266	2.734679	0.92	0.6205	1.0949
2.1.6	1	1	1	1	1
2.2.4	1.03954	1.269908	0.893	0.9405	1.333
2.2.5	1	1	1	1	1
2.2.6	1	1	1	1	1
2.3.4	0.9878	2.5809	0.92	0.6579	1.0872
2.3.5	0.96263	2.724583	0.92	0.6198	1.0960
2.3.6	0.89423	2.75	0.92	0.62037	1.0951
3.1.1	0.80162	1.4071	0.92	0.6983	1.0501
3.1.2	1.54745	1.314	0.96	1.2766	1.5515
3.1.3	1.21551	1.106	0.98	1.2155	1.2155
3.1.4	1.05	1.05	0.98	1.05	1.05
3.1.5	1.05	1.05	0.98	1.05	1.05
3.1.6	1	1	1	1	1
3.2.1	0.98042	2.584	0.92	0.6591	1.0450
3.2.2	1.04738	0.954	0.85738	1.0474	1.1573
3.2.3	1.21551	0.966	0.81451	1.2155	1.2155
3.2.4	0.9025	1.1025	0.92	0.9025	1.0497
3.2.5	1	1	1	1	1

3.2.6	1	1	1	1	1
3.3.1	0.9580	2.6	0.92	0.6554	1.0525
3.3.2	1.06677	2.342	0.94	0.7256	1.0380
3.3.3	0.97872	2.544	0.92	0.6696	1.0430
3.3.4	1.02784	2.4624	0.94	0.6904	1.0450
3.3.5	0.9798	2.59	0.92	0.6576	1.0508
3.3.6	0.95306	2.588	0.92	0.6582	1.0503
4.1.2	1.6094	1.568	0.98	1.0752	1.4410
4.1.3	1.42269	1.748818	0.96	0.9522	1.2675
4.1.4	0.95	1.05	0.98	0.9588	1.05
4.1.5	1	1	1	1	1
4.1.6	1	1	1	1	1
4.2.2	2.43829	1.0469	0.94	1.5939	2.1055
4.2.3	2.10936	1.088537	0.9	1.5207	2.006
4.2.4	0.95	1.05	0.98	0.9613	1.05
4.2.5	1	1	1	1	1
4.2.6	1	1	1	1	1
4.3.2	1.41224	1.782	0.96	0.9480	1.2619
4.3.3	0.97723	1.276282	0.912	0.92996	1.2173
4.3.4	1	1	1	1	1
4.3.5	1	1	1	1	1
4.3.6	1	1	1	1	1
5.1.3	1.97993	1.1613	0.95	1.4364	1.8796
5.1.4	1.21551	1.1802	0.90024	1.2155	1.2155
5.1.5	1.6289	1.216	0.84	1.3769	1.6289
5.1.6	1	1	1	1	1
5.2.3	2.1829	1.108	0.987	1.5081	1.9943
5.2.4	1.1576	1.1576	0.92	1.1576	1.1576
5.2.5	1	1	1	1	1
5.2.6	1	1	1	1	1
5.3.3	1.1576	1.142	0.94	1.1576	1.1576
5.3.4	1.88565	1.116	0.98	1.4984	1.8856
5.3.5	2.48975	1.1718	0.931	1.4248	1.8859
5.3.6	2.29915	1.106	1	1.5131	1.9997

Chapter 7

Conclusions

Chapter 7: Conclusions

7.1 Key Findings

This thesis presents a new way to find the minimal TAC of HENs and MENs that are designed in detail and makes use of a combination of methodologies in a novel optimisation framework. The key findings are summarised below:

1. Current MINLP optimisation techniques provide a useful way of finding optimal networks for HENs and MENs, however the current solver technology fails to guarantee globally optimal solutions, particularly for large problems. For this reason the current formulations make use of shortcut models for the individual units within the networks. Through the inclusion of implicitly-determined, match-specific corrections that force the MINLP topology optimisation toward the actual, detailed, rigorously-determined designs it is possible to represent detailed information about these designs in the MINLP without increasing the non-convexity of the model. In this way it is possible to use information from detailed models to update the MINLP topology optimisation in such a way so as to generate networks that take this information into account.

2. By utilising these correction factors in the MINLP it is possible to generate networks that have taken into account many of the details of a design that have not previously been included at the level of network design. For HENS this information includes the F_T correction factor, the number of shells, the actual log mean temperature difference, rather than an approximated one, the pressure drops, overdesign, changes to the overall heat transfer coefficient that are dependent on velocities and fluid properties, and the TEMA designs. For multi-period HENS it is possible to consider the addition of extra exchangers that may be necessary in some periods due to vastly different heat demands in different periods, as well as the changes to heat transfer coefficients in different periods as a result of differing velocities and temperatures. In MENS the detailed mass exchange fluxes along the column can be

considered through the use of corrections to the overall height, as well as flooding considerations, the selection of the size of packing and the resulting effect the size of the packing has on the cost of each individual exchanger. All of these considerations that are vitally important in the design of the networks have not previously been considered at the network design level, rather having been calculated after the network is determined. These inclusions allow for the MINLP to determine the optimal network while still considering the details of the individual units.

3. Even though shortcut models are used to represent the individual units in MINLP network optimisation, the formulations are still non-convex. This results in great difficulty in finding globally optimal solutions, especially for larger problems. By limiting the change of the correction factors between each iteration in the algorithm and by selecting initial parameters that underestimate the objective function, it is possible for many candidate networks to be evaluated during the course of the algorithm, with each network designed in detail after the MINLP optimisation to validate the solutions obtained. By providing the MINLP with numerous initial points as a result of the changes in correction factors, the chances of obtaining a globally optimal solution are greatly enhanced. In other methods that have been used there has not been a rigorous and systematic method proposed that provides different initial points in order to find an optimal solution. The methodology presented in this thesis, not only provides a systematic method of generating many initial points, it also ensures that the networks that result from these optimisation are validated and checked using detailed models.

4. Previous authors have not attempted to validate the designs obtained at the MINLP level, resulting in networks that could not be shown to be optimal once all of the detailed design considerations were taken into account. Methods to date would use the MINLP optimisation to find the optimal network topology and use the heat and mass balances to design the detailed units. Without knowledge as to whether the network is globally optimal or whether additional information from the detailed designs would change the solution drastically. The method presented in this paper rigorously determines the detailed network after the MINLP optimisation, in doing this it is clear that the shortcut models required in the MINLP optimisation fail to

properly account for many aspects of a design that significantly affect the final costs involved. The use of the validation step is vital in finding optimal networks. In addition, saving the detailed designs between each run of the MINLP and using these detailed networks to determine the optimal network is of extreme importance. Previous authors only used the solution of the MINLP objective function to determine optimal networks, whereas the method presented in this thesis uses the rigorously determined designs as the key determination of the optimal network.

5. In addition to the novel methodology that was presented for the synthesis of HENs and MENs, a new method for the determination of optimal packed columns was developed. Orthogonal collocation on finite elements (OCFE) was successfully implemented to optimise an entire network of columns simultaneously once the topology, mass balances, and split flows, were determined through the use of an MINLP optimisation. The novel OCFE model was used to determine the optimal packing sizes, diameters, and heights of the packed columns based on a novel formulation that found fast and reliable solutions for a variety of problem sizes.

7.2 Future Work

As described above, the methodology presented here lays a general framework for the synthesis of more realistic and, as was shown, often optimal mass and heat exchanger networks. This general framework has been applied to a number of applications in Chapters 4, 5, and 6. The methodology itself can be potentially improved through the following investigations.

7.2.1 Algorithmic alterations

By decreasing the maximum change that a correction factor can undertake between successive iterations it may be possible to find even more potential networks, further increasing the efficacy of the method in two ways. Firstly, by having more initial conditions that allow for more restarts, it is more likely that a globally optimal solution is found at some

point throughout the iterations. The second improvement that this would result in is that fewer potential solutions will be excluded as the solution space is shifted through the changing parameters in smaller increments, potentially also increasing the chances of convergence of the solutions between the NLP and MINLP. While these may result in improvements to the results, the methodology will take a lot longer to converge to solutions; a potentially large setback, particularly for its application to the individual exchanger designs that are done manually, as was the case in the HENS examples.

Furthermore, since a major problem associated with this methodology lies in the formulation and solution of the MINLP, improvements need to be made to the way in which the equations are formulated so as to still include non-isothermal into the HENS MINLP and find feasible solutions that are globally optimal with current solver technology. Otherwise, improvements in MINLP solver technology need to be made so as to allow the current formulations to be solved in a more fast and reliable way to globally optimal solutions.

7.2.2 HENS extensions

In Chapter 4 the methodology was applied to HENS, with new features that have yet to be included in HENS included through the inclusion of correction factors. A number of minor additions to the model could enhance the work in the future.

A new formulation for pressure drops could be included so that the pressure drops are in some way related to the size or type of exchanger present. To date little work has been done on including pressure drops into HENS, and the task is made more complex by the fact that the pressure drop is strongly related to the number of tube and shell passes present in each individual exchanger. It is of the opinion of the author that the implicit inclusion of pressure drop in this study could be further enhanced through the development of a more detailed and nuanced approach to pressure drops whereby the MINLP has access to some sort of explicit correlation. This correlation could potentially add to the non-convexity of the problem, however it may be possible to develop a simplified correlation that will increase the usefulness of the approach.

Further extensions that can be made to the work can be to include more complex aspects that may be present in real industry examples. This can involve phase changes for the process streams, the optimisation of the different levels of steam required for the utilities,

or the inclusion of multiple exchanger types through the use of disjunctions. It can also be extended easily to include different superstructure approaches, such as the hyperstructure approach of Floudas and Ciric (1986) and the match-centric approach of Huang and Karimi (2012).

The method chosen for the optimisation of the individual heat exchangers for Chapters 4 and 5 was the industry-standard heuristic approach that utilises a number of empirical correlations and rules-of-thumb. During Chapter 4 and 5 it is mentioned a number of times that a different approach can easily be utilised at this step of the algorithm to make up for the fact that the process of designing the network in detail can be a lengthy process using this method. It is possible to automate this process through the use of a sub-optimisation, like the NLP approach used by Mizutani, et al. (2003a) and the MINLP formulation of Ravagnani and Caballero (2007a). These methods will greatly increase the speed of the algorithm however, as was noted during the chapter, they increase the chances of local optima. In addition to this, the method will probably require numerous restarts as the MINLP solvers can find it difficult to find feasible solutions in such large problems. It would be interesting to use these suboptimisations in an automated procedure using the algorithm of this study, rather than their own, to compare solutions. In addition to these models, it may be interesting to use some commercial software that use rigorous thermodynamic models for the modelling of the individual units, as opposed to these heuristic methods. While these commercial packages (such as ASPENTECH's HTRI™) are often costly they would provide high-quality solutions that can be used to validate and compare the designs obtained using the various methods.

While these more automated methods may be utilised in the single period problems it may be difficult to implement them in the multi-period problems covered in Chapter 5. Many engineering judgement decisions were required during the design of the multi-period networks, therefore it may be an interesting extension to design an MINLP decision-making tool to optimise the multi-period individual units. This MINLP may need to have numerous disjunctions built-in that may lead to large problems that will be difficult to solve to global optimality.

Further improvements to the multi-period formulation that may be able to be more easily implemented in the current framework could be the inclusion of bypasses in the individual exchanger designs to avoid the use of extra exchangers in some periods, and to minimise the overdesign of the units in certain others. Furthermore, pressure drop considerations could be included in a manner similar to that included in the single-period paper.

7.2.3 MENS extensions

Chapter 6 shows the application of the algorithm to MENS, concentrating on packed columns in examples with single components and without regeneration. Obvious extensions that future studies can undertake should involve the inclusion of regeneration of the absorbents, as well as the inclusion of examples with multiple components or with reactive mass exchange. Since MENS is generally an understudied area of process integration, there is a lot of room for expansion in regards to these. In addition, the novel NLP suboptimisation developed in the study can be further extended to select different packing types during the optimisation through the inclusion of disjunctions.

If disjunctions are included it may also be possible to extend the work to include staged columns by utilising MINLP suboptimisations, or by using some of the shortcut models developed by Kamath, et al. (2012) for the optimisation of distillation columns.

By extending the NLP suboptimisation of this study to include more detailed aspects of packed column design, such as temperature and thermodynamically linked methods for the calculation of mass transfer coefficients it may also be possible to include the optimisation of combined heat and mass exchanger networks (CHAMENs) within the optimisation framework presented in this thesis.

While the novel NLP suboptimisation developed in this study is very flexible and allows for the method to be automated, an interesting study could be conducted by modelling the individual packed columns within a variety of different frameworks and comparing and validating the solutions obtained. Through using different packages such as HYSIS, ASPEN, and commonly used HETP methods, it can be determined which methods are most suitable and in what ways the method of this study can be improved.

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8. References

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Nomenclature

9. Nomenclature

Abbreviations

ACC	-	annualised capital costs
BCC	-	balanced composite curve
BCCC	-	balanced cold composite curve
BHCC	-	balanced hot composite curve
CCC	-	cold composite curve
CHAMENS	-	combined heat and mass exchange networks
DAE	-	differential algebraic equation
DE	-	differential equation
DEM	-	differential evolution method
DFP	-	driving force plot
DICOPT	-	discrete and continuous optimiser
EA	-	evolutionary algorithm
EMAC	-	exchanger minimum approach composition
EMAT	-	exchanger minimum approach temperature
GA	-	genetic algorithm
GAMS	-	General Algebraic Modelling System
GBD	-	generalised Bender's decomposition
GDP	-	generalised disjunctive programming
HCC	-	hot composite curve
HE	-	heat exchanger
HEN	-	heat exchanger network
HENS	-	heat exchanger network synthesis
IBMS	-	interval based MINLP superstructure

I-O	-	input-output
IIP	-	integer infeasible path
IVP	-	initial value problem
LCC	-	lean composite curve
LMTD	-	logarithmic mean temperature difference
LMCD	-	logarithmic mean composition difference
LP		linear programming
MEN	-	mass exchanger network
MENS	-	mass exchanger network synthesis
MILP	-	mixed-integer linear program
MINLP	-	mixed-integer nonlinear program
MSA	-	mass separating agent
NLP	-	nonlinear program
NPV	-	net present value
OA	-	outer approximation
OA/ER	-	outer approximation/equality relaxation
OA/ER/AP	-	outer approximation/equality relaxation/augmented penalty
ODE	-	ordinary differential equation
PT	-	pinch technology
PTA	-	problem table algorithm
RCC	-	rich composite curve
SA	-	simulated annealing
SBB	-	simple branch and bound
SBS	-	supply based superstructure
S&TBS	-	supply and target based superstructure
S&THE	-	shell and tube heat exchanger
SQP	-	sequential quadratic programming
TAC	-	total annual cost
TEMA	-	Tubular Exchanger Manufacturers' Association
T&SBS	-	target and supply based superstructure

Symbols

ΔT_{min}	-	minimum approach temperature (K)
$\Delta y_{lm,k}$	-	logarithmic mean composition difference
A	-	removal factor
A	-	area (m^2)
a	-	fixed cost of an exchanger
a	-	interfacial area of packing
b	-	cost coefficient
c	-	cost coefficient
C_j	-	cold stream j
$CC_{per\ unit}$	-	capital cost of a single unit
E_M	-	Murphree efficiency factor
G_i	-	flowrate of the gaseous stream
h	-	Individual stream heat transfer coefficient
h_j^*	-	cost weighted individual stream heat transfer coefficient
h_i	-	length of finite element
H_{min}	-	minimum height
H_i	-	hot stream i
k	-	temperature location
K	-	number of collocation points
ky	-	overall mass transfer coefficient
L_j	-	flowrate of the liquid stream
$LMTD$	-	logarithmic mean temperature difference
m_j	-	slope of the equilibrium line
$ncol$	-	number of collocation points
ne	-	number of finite elements
N	-	flux

$N_{min\ units}$ - minimum number of exchangers

N_{real} - number of stages at non-equilibrium

$N_{streams}$ - number of streams

q - enthalpy exchanged (kW/s)

$T_{H1,s}$ - supply temperature of hot stream 1

$T_{C1,s}$ - supply temperature of cold stream 1

$T_{H1,t}$ - target temperature of hot stream 1

$T_{H1,t}$ - target temperature of hot stream 1

U - overall heat transfer coefficient

U_{min} - minimum number of heat exchangers

x - rich stream composition

x - continuous variables

XSA - cross sectional area

y - lean stream composition

y^K - binary variables

y^* - equilibrium composition of contaminant in lean stream

z - height along a packed column

Ω_q - interpolating polynomial

Φ_j - weighting factor for non-uniform exchanger specifications

It should be noted that Chapters 4, 5, and 6 each have their own nomenclature and if any symbols and abbreviations are missing from this section it is likely that they are included in the relevant chapters.